DOE Division of Materials Sciences & Engineering
Condensed Matter Physics & Materials Chemistry
Program Review

Ames Laboratory
Ames, Iowa

May 7-8, 2001

Program Contacts:

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AMES LABORATORY

PROGRAM REVIEW AGENDA

For

CONDENSED MATTER PHYSICS
&
MATERIALS CHEMISTRY

MAY 7–8, 2001

Monday, May 7

8:00	Reviewers meet with DOE
8:15	Welcome. Overview of Ames Lab/ISU—Tom Barton, Director
8:25	Overview of Materials Sciences—David Hoffman, Associate Director
8:35	Overview of Materials Chemistry—Pat Thiel, Program Director
8:50	Overview of Condensed Matter Physics—Bruce Harmon, Program Director
9:05	New Materials—Paul Canfield
9:35	Posters for New Materials
11:45	Lunch (Catered for reviewers)
12:45	Overview Talk for Session A
1:05	Posters for Session A
3:10	Organization for tours and one-on-one discussions
3:20	Tours of facilities (reviewer's choice)
4:00	One-on-one discussions
6:00	Evening (To be announced)

Tuesday, May 8

8:30	Caucus (Reviewers ask questions of management or make requests)
9:00	Quasicrystals—Pat Thiel
9:20	Magnetic Molecules—Marshall Luban
9:40	Posters
11:40	Overview Talk for Session B
12:00	Lunch + Posters for Session B
2:15	Wrap-up one-on-one discussions
3:30	Close out (Reviewers with DOE, Management)

Overview

For more than 50 years the Ames Laboratory has been internationally recognized for forefront materials research. While the methods, facilities, and materials have often changed, the tradition and dedication to a world class effort remains sacrosanct. We welcome this opportunity to present our current science projects, and we hope to do so in a way that captures at least part of the overall community that is the Ames Laboratory.

The Condensed Matter Physics and the Materials Chemistry programs are being reviewed together. Since many of the projects involve researchers from both programs (and also from the Metallurgy and Ceramics program) the review is not separated into two parts, but rather into poster sessions grouped by scientific themes. The poster sessions are to be the heart of the review with the purpose of having the reviewers interact with scientists one-on-one. As the emphasis of the review is personal interactions between reviewers and scientists, we have included in this booklet single page descriptions of the research highlights of the principal scientists, followed by a list of their group's publications since the last DOE program review (fall 1998). While these pages highlight the breadth and quality of the personal contributions, the real strengths of the laboratory come to the fore when teams of researchers collaborate and push themselves to achievements well beyond the sum of the individual efforts. Indeed, collaborative teams help define the Ames Laboratory. To capture some of that flavor we have somewhat arbitrarily divided the overall research into 8 scientific themes and included a brief one-page description for each. Condensed Matter Physics and Materials Chemistry programs are not separated in these theme pages. To help sort out the administrative organization of the programs, we include the breakdown of projects as submitted in the Work Authorization System.

The Budget for the two programs for the present fiscal year is:

- Materials Chemistry: $2,079k,
  Chemical Structures: $454k, Engineering and Polymer Chemistry: $793k, High Temperature and Surface Chemistry: $832k
- Condensed Matter Physics: $4,930k.
  Neutron and X-ray Scattering: $1,086k, Experimental Condensed Matter Physics: $2,507k, Theoretical Condensed Matter Physics: $1,337k

Unlike previous reviews, we have not included any reprints or preprints of journal articles. Reviewers who would like this information can obtain pdf files by going to the web http://cmpweb.ameslab.gov/programreview/2001/

Condensed Matter Physics Office Number: (515) 294-3481
Materials Chemistry Office Number: (515) 294-7568

Prologue

Laboratory Mission:

> The Ames Laboratory will conduct fundamental research in the physical, chemical, materials, and mathematical sciences and engineering which underlie energy generating, conversion, transmission and storage technologies, environmental improvement, and other technical areas essential to national needs, . . .

BES Mission:

> The mission of the Basic Energy Sciences (BES) program is to foster and support fundamental research in the natural sciences and engineering to provide a basis for new and improved energy technologies and for understanding and mitigating the environmental impacts of energy use.

Given the above two mission statements, it is not surprising that you will find our scientists engaged in fundamental research. What is not stated explicitly in either statement is the expectation that the research be conducted at the highest level. It is an expectation we strive for as scientists and as an institution, and we expect to be reviewed by such standards.

While the various Ames Laboratory groups more than hold their own going after "complex materials" or "nano-scale materials," the primary motivation driving the science in Ames comes from smart people fascinated by learning, and driven by curiosity. Photonic Band Gap materials sprang from local ideas, and expanded because of funding opportunities. We are grateful for the atmosphere that permits new ideas to flourish.

Program and Group Organization
Condensed Matter Physics & Materials Chemistry
(From the Work Authorization System)

This booklet presents examples of team efforts and then highlights the individual scientists, each of whom may be working in many projects, so it might be hard for a reviewer to actually know what to review. A good rule is to “follow the money,” which comes into the programs via a Work Authorization System. The different projects listed below are broken out according to this year’s WAS. The total dollar amounts for the sections are given in the Overview.

I. CONDENSED MATTER PHYSICS

A. Neutron and X-ray Scattering

Neutron Scattering
C. Stassis, D. Vaknin, J. Zarestky

X-ray Scattering
A. I. Goldman

MUCAT
A. I. Goldman

Bending Magnet Beamline
A. I. Goldman

B. Experimental—Condensed Matter Physics

Optical and Spectroscopic Properties of Solids and Surfaces
D. W. Lynch, C. G. Olson, M. C. Tringides

Superconductivity
D. K. Finnemore, J. E. Ostenson

New Materials and Phases
F. Borsa, D. C. Johnston, L. L. Miller, C. A. Swenson

Semiconductor Physics
J. Shinar

Correlated States in Magnetic Materials
P. C. Canfield, A. I. Goldman, B. N. Harmon, D. W. Lynch, R. Modler, C. Stassis

Photonic Band Gaps
R. Biswas, K. Constant, K.-M. Ho, C. M. Soukoulis, G. Tuttle

C. Theoretical Condensed Matter Physics

Optical and Surface Physics
K.-M. Ho, C. Z. Wang

Superconductivity
J. R. Clem, V. G. Kogan

Electronic and Magnetic Properties of Solids
B. N. Harmon, K.-M. Ho, M. Luban, J. H. Rose, J. Schmalian, C. M. Soukoulis

Computational Materials Sciences Network (CMSN)
B. N. Harmon

II. MATERIALS CHEMISTRY

A. Chemical Structures

Structural and Electronic Properties of New Main-Group Intermetallic Phases
J. D. Corbett

Zintl (Valence) Compounds in Intermetallic Systems and Their Stabilization by Interstitial Atoms
J. D. Corbett

Electronic and Atomic Structure of Quasicrystals and Closely-Related Phases
G. J. Miller

B. Engineering and Polymer Chemistry

Manipulating Chemistry through Materials Processing
M. Akinc and A. J. Thom

Synthesis and Study of Organometallic Monomers and Polymers as Preceramic and Optoelectronic Materials
T. J. Barton

New Smart Environmentally Sensitive Polymers and Polymer-Quasicrystal Composites
S. Mallapragada

New High Performance Polymers and Polymer/Quasicrystal Composites
V. V. Sheares

Solid-State NMR of Heterogeneous Polymer Systems
K. Schmidt-Rohr

C. High Temperature and Surface Chemistry

New Initiative on Surface and Interface Properties of Quasicrystals
P. A. Thiel

Electrocatalytic Materials for Oxygen-Transfer Reactions
D. C. Johnson

Scientific Themes

I. Neutron and X-Ray Scattering

Thermal neutrons have wavelengths and energies ideally matched to the structure and frequencies of crystals and their elementary excitations. Neutron scattering studies have been instrumental in elucidating the basic science of hundreds of new materials and crystals first developed at the Ames Laboratory. Historically, it is the study of rare earth magnetism that first established the neutron scattering tradition at Ames. This work was carried out in the 1950s by collaboration between Ames Lab and Oak Ridge scientists. In the '60s and '70s, Ames Lab operated its own research reactor and a strong on-site tradition in neutron scattering developed. With the shut down of the Ames reactor, our instruments were moved to ORNL, where we established the first neutron scattering user group in the U.S. Nowadays, our neutron scattering studies are mainly conducted at the High Flux Isotope Reactor (HFIR) at ORNL, although facilities at IPNS (Argonne), NIST, and in Europe are also utilized. Of particular importance is the HFIR HB1-A spectrometer which was designed, constructed, and now operated as a user's facility jointly by the Ames and ORNL neutron scattering groups. Jerel Zarestky is the Ames Laboratory scientist, permanently located at Oak Ridge, who oversees the maintenance, upgrades, and operation of the instrument, in addition to his involvement with scientific investigations. Also, an Ames Laboratory triple-axis spectrometer was installed at the U. of Missouri Reactor, has been modernized and is in operation. Costa Stassis is the senior scientist overseeing the neutron scattering effort, with some of his time devoted to x-ray scattering, and David Vaknin is a principal investigator equally using neutron and x-ray scattering techniques.

With the rapid development of national synchrotron facilities, modern x-ray scattering methods have become an essential component of any serious materials laboratory. Alan Goldman has led the Ames Laboratory's involvement. He is director of the Midwest University Collaborative Access Team (MUCAT) which has built and is operating an undulator beam line at the Advanced Photon Source. A side station has recently been built with collaboration and financial support from Jülich, and funding and construction has just recently started for an adjacent bending magnet beam line. Research facilities for magnetic x-ray scattering are completed and several key experiments have been performed. The students in Dr. Goldman's group are frequently exposed to complementary neutron scattering techniques when the science demands it and the opportunities arise. David Vaknin has recently commissioned a custom built liquid surface reflectometer at the MUCAT beamline. In collaboration with scientists in the Metallurgy and Ceramics division, Alan Goldman has recently commissioned a unique powder defractometer furnace (temperatures up to 1600°C), which allows rapid (seconds) monitoring of structural changes in materials.

The neutron and x-ray scattering groups are among the most collaborative in the Ames Laboratory. The unique information available from these techniques is absolutely essential for elaborating the physics of many systems, a fact not lost on the many other Ames Lab groups that either have new samples to study or new theories to test. A glance at the publication lists of these scientists reveals how broadly their interactions are spread.

Also worth noting are the x-ray laboratories existing on-site at the laboratory. There are three rotating anode facilities (Goldman, Vaknin, Gschneidner, Jr.), and numerous more standard x-ray units (Johnston, Goldman, Chumbley (2), Cook, and Lograsso (2), Corbett, and Miller).

II. Optical and Spectroscopic Properties of Solids and Surfaces

Measurements determining the energy states and elementary excitations have traditionally been the pathway to understanding the electronic structure and physical properties of materials. In this synopsis, we focus on several of the spectroscopies which are key to the characterization of solids and surfaces. Angle-resolved and angle-integrated photoelectron spectroscopies are used to study the electronic structure of metallic solids with expected correlated-electron effects. Monochromatic photons in the 5-800 eV range are used so both valence electrons and core electrons may be accessed. This is carried out at our Ames Laboratory beam lines at the Synchrotron Radiation Center, Stoughton, Wisconsin. Cliff Olson is the Ames Laboratory scientist located at Stoughton who designs, builds, maintains, and uses the equipment for high resolution studies. Cliff and Dave Lynch were the first group to use photoemission to measure the band gap opening in high T_c superconductors. They have recently published a comprehensive monograph, *Photoemission Studies of High-Temperature Superconductors* (Cambridge University Press, 1999). This group has strong collaborations with outside groups and with experimentalists and theorists within the Ames Laboratory.

For surface studies, High Resolution LEED provides information about the average morphology of the surface from the sharpness of the diffraction spots and Scanning Tunneling Microscopy (STM) provides local real space information about the grown structures. Michael Tringides' emphasis has been on studies of growth mechanisms (surface diffusion, nucleation, island formation, atom ordering, etc.), and recently on the mechanisms giving rise to uniform height nano-scale islands observed during the growth of Pb/Si(111). Collaboration with theorists K.-M. Ho and C.-Z. Wang have led to quantum size effect explanations. Photoemission measurements are being made to look for quantum well levels in these islands.

Measurements of the optical conductivity (diagonal and the magneto-optical off-diagonal components) are made from 0.5 to 5eV in Dave Lynch's lab, where temperatures down to 2K and fields up to 7 T can be applied. There is close collaboration between this group and the magnetic structure theorists (Harmon, Antropov) since the correlated electronic states encountered in magnetic materials often require approaches beyond the standard LDA band structure methods. Magneto-optics is sensitive to such states.

Neutron and x-ray spectroscopies are described in summary I. The Auger and TEM spectroscopies at Ames Laboratory are primarily supported and utilized through collaborations with scientists in the Metallurgy and Ceramics program. For about ten years, a commercial X-ray photoelectron spectrometer has been made available for users in the Lab (free of charge) by the Materials Chemistry program. It has proven invaluable for identification of oxidation states and for determining the surface composition of materials.

III. New Materials, Phases, and Phenomena

To some extent the entire materials sciences program could be fit under this category, so the description here is arbitrarily selective and by necessity brief. A few recent or classic examples are given to characterize the program. The Ames Laboratory's tradition in the synthesis and characterization of new materials can be traced back to the Manhattan project, when over 2 million pounds of pure uranium metal were produced, based on "the Ames process." Materials can be synthesized, chemically analyzed, and physically characterized in a remarkably short period by making use of scientists in all parts of the laboratory. Because of Frank Spedding's fondness for rare earths, magnetic materials have a special niche and are described separately in summary IV. Likewise, quasicrystal research has broadly developed and is described in summary VI.

The synthesis of new compounds with unusual bonding character and often exotic crystal structures has long been the forte of John Corbett and other Ames Lab solid state chemists. Following some studies of Dr. Corbett, Mufit Akinc's group investigated interstitially modified titanium silicide and discovered remarkable high temperature oxidative stability and other desirable properties. Related theoretical studies by K.-M. Ho's group looked at bonding in molybdenum silicides, and groups at ORNL and elsewhere have pursued the materials for applications. For some time Doug Finnemore has screened nearly all samples from the chemistry groups for unusual low temperature behavior. Unfortunately, MgB_2 was never one of the samples; but once word reached Ames of the superconductivity in this material, Paul Canfield and Doug Finnemore had a field day with three *Physical Review Letters* published in March. At the same time, Vladimir Antropov (and collaborators at NRL) submitted a PRL on the theory (out May 7). At least 12 more Ames papers have been published or are in process. Superconductors and related compounds continue to be a strong focal points also for David Johnston and Ferdinando Borsa, both of whom have extensive collaborations. John Clem and Vladimir Kogan represent perhaps the strongest two theorists in the phenomenology of type II superconductors, and they collaborate readily with experimental groups.

There are many other systems that have been examined by various subsets of these researchers: LiV_2O_4 (oxide heavy ferimion), RNi_2B_2C (magnetic supreconductors), RMgZn (magnetic quasi-crystals), $AlMgB_{14}$ (hard material).

Facilities to pursue forefront materials research are essential. It has often been remarked that there are more Quantum Design units (7) in Ames than at their factory in San Diego. There are also many custom built units, such as the dilution refrigerator (Modler), NMR spectrometers (Borsa), optical and electron spectrometers (Lynch, Olson), and specific heat and thermal expansion units (Swenson). Perhaps most impressive are all the facilities for materials synthesis. Besides the specialized world class facilities available at the Lab's Materials Preparation Center, facilities for thin film and crystal growth abound in individual groups (Canfield, Johnston, Shinar, Miller, Corbett).

IV. Magnetism (Materials, Molecules, Modeling)

Frank Spedding's work on rare earth chemistry led to the local war time effort on uranium ("If you understand 4f elements you can be our expert on 5f's."). After the war and the establishment of the Ames Laboratory (1947), research on rare earth metals and compounds resumed in earnest, now with strong interactions among the chemists, metallurgists, and physicists. Frank Spedding held joint appointments in all three academic departments and hired a postdoc in each area to work under his supervision. Good traditions are to be treasured and exercised whenever possible, and the magnetism expertise that has evolved over the years is one of the cornerstones of the Ames Laboratory's international reputation. Only a few recent examples are given.

The RNi_2B_2C family of magnetic superconductors is an example of what can be accomplished when materials synthesis, detailed measurements of physical properties, and theoretical expertise are combined. Within a few months of the discovery of the RNi_2B_2C materials (Cava et al.), researchers at Ames were able to grow high purity single crystals of this quaternary superconductor. This ignited a large experimental and theoretical effort to understand the magnetic and superconducting interactions. Specifically, a combination of Fermi surface effects as well as CEF splittings greatly affect the superconducting state (Bud'ko). Neutron scattering experiments (Stassis) studied the superconducting effects on phonon softening and line widths as well as the subtleties in the phase transitions of the vortex lattice (Kogan, Lucent, Risø). NMR (Borsa) investigations confirmed that $YbNi_2B_2C$ is indeed a correlated electron metal and magnetic X-ray scattering (Goldman, Detlefs {ESRF}) showed the wave vectors of the ordered state vary with temperature and compare well with band structure calculations (Harmon).

Other compounds that have been examined by several groups of Ames Lab researchers are local moment systems such as RNi_2Ge_2 and $RAgSb_2$ (magnetic phase diagrams and met magnetism) as well as RMgZn and $(Y_{1-x}Tb_x)Ni_2Ge_2$ (model spin glass systems with periodic and anisotropic local moments, respectively).

Recent advances in synthesizing large and interesting magnetic molecules and their single crystals are allowing exploration of the bridge between microscopic, atomistic magnetism and the complex interactions and cooperative phenomena encountered in larger magnetic structures. Phenomena such as magnetic tunneling and quantum coherence have motivated F. Borsa (NMR), R. Modler (low T), C. Stassis (inelastic neutrons), L. Miller and D. Vaknin (thin films), D. Lynch (optics), A. Goldman (X-rays), Canfield (temperature and field dependent magnetization and specific heat), M. Luban and B. Harmon (theory).

$Gd_5Si_2Ge_2$ is another example of a remarkable rare earth compound that is being studied by researchers in all three programs. It undergoes a martensitic phase transition from a high temperature orthorhombic phase to a ferromagnetic monoclinic phase, which can be driven by applied magnetic fields. It has the highest magneto-caloric effect of any known material, and with a Curie temperature near room temperature it is being developed (outside the BES program) for magnetic refrigeration applications. Karl Gschneidner, Jr. and Vitalij Pecharsky (Met. and Cer.) lead a team to synthesize and characterize these materials, which also exhibit huge magneto-elastic effects and giant magneto-resistance. Gordon Miller is investigating structure and bonding, while Alan Goldman and Costa Stassis are involved with magnetic x-ray and neutron scattering. Theorists (Harmon and Antropov) are performing first principles calculations. These are complex materials (36 atoms/cell).

V. Photonics

The Ames Lab group lead by Kai Ming Ho and Costas Soukoulis has played a dominant role in developing the field of Photonic Band Gap (PBG) materials. In fact, their calculation showing that PBGs with diamond (or diamond-like) lattices have 3D band gaps kept the field from withering away at a time when there was considerable pessimism about the viability of PBGs in general. After that, the field has exploded, with over 500 groups worldwide having web pages on the topic. The basic PBG idea is to design new structures with periodically modulated dielectric constants, thereby influencing the properties of photons in much the same way ordinary solids influence the properties of electrons. PBG materials have immense potential for creating a new class of optical devices, which, if realized, could result in optical circuits and communications devices as powerful and versatile as electronic circuits. With PBG materials, one can make resonant cavities for light amplification, suppress unwanted optical losses and make light execute the sharp turns needed for circuit miniaturization.

Ames Lab experimentalists (G. Tuttle, K. Constant, W. Leung) joined the effort in 1993, and have investigated fabricating PBGs using novel techniques, from microfabrication using Si wafers to fcc ordered nano-meter sized air holes in TiO_2 ceramics. Rana Biswas has been exploring ways (both computationally and experimentally).that PBGs can greatly enhance directional antenna performance. Alan Goldman has studied the nano-structured PBG ceramics with X-ray scattering, and both Joe Shinar and David Lynch are investigating optical properties of PBGs under special conditions.

Besides purely ceramic PBGs, metallic PBGs of very small filling ratios (volume of metal to the total PBG volume) offer a number of potential advantages, in particular their ease of fabrication, their lightness and their flexibility and strength. These novel PBGs are being investigated computationally using transfer matrix and finite difference time domain (FDTD) algorithms to understand their properties and to help design structures with unique properties.

Left-handed materials (LHM) is a very new area of photonics research being studied in Ames (Soukoulis). LHM have both the dielectric constant and magnetic permeability negative. Although there are no naturally occurring materials with $\mu < 0$, these can be achieved by artificially fabricated structures. LHM have unique and potentially useful properties, as many basic EM phenomena, such as the Doppler shift, Snell's law, and Cerenkov radiation are "reversed." The powerful FDTD techniques developed for PBG research are being used to understand the very interesting, challenging, and mostly unexplored properties of LHM.

Another aspect of photonics is being investigated by Joseph Shinar. His group explores the mechanisms affecting the light emission in organic materials (mostly polymers). By choosing to look at interesting blue light emitting diodes, their basic research has led to a discovery of a blocking mechanism to prevent electron hole recombination, and it also led to a scheme to overcome the blocking. Needless to say, blue LEDs are unusual, but perhaps not so in the near future thanks to this work.

VI. Theory, Simulation, Computation

Theory and experiment are perhaps best classified as approaches to science. Simulation or computation is now regarded by many as a third approach, although it is perhaps better to think of computing as a remarkable enabling tool for both theory and experiment. All of the "theorists" at Ames Laboratory are deeply engaged with experimental groups, and certainly computers are essential to both. It is not unusual to have a graduate student split between theory and experiment, with his/her thesis containing extensive comparison between experiment and calculated results. Computing beyond ordinary PCs is also a big deal. Iowa State U. lays claim to the development of the first electronic digital computer, invented by John V. Attanasoff, a Physics professor motivated to help his students solve quantum mechanics problems in 1937-40. We are still following that tradition. In partnership with scientists in the Applied Mathematics program (e.g., Dave Turner), a large number of cost effective "almost supercomputing" clusters have been built, and codes have been parallelized to run on them. For much larger supercomputing needs, the facilities at NERSC or at ORNL are utilized via a T3 line.

Among the codes recently developed, tight binding molecular dynamics (Ho, Wang) has been adapted by a large number of groups worldwide. Its impact resulted in a Materials Science Award for Outstanding Sustained Research. Spin Dynamics (Antropov, Harmon) is more recent, but it too has been adapted by a number of groups and has been employed by collaborators at ORNL to win the Gordon Bell prize for the fastest application (sustained one TeraFLOPS). Codes to solve the full Maxwell’s equations for photonic band gap materials (Ho, Soukoulis) were first developed in Ames in 1990, and the field has exploded a hundred-fold since.

Although also using computers, John Clem and Vladimir Kogan are more traditional, using strong analytical skills to elucidate aspects of type II superconductors. They have been particularly active with high temperature superconductors. Marshall Luban also combines keen analytical skills with computing, when going after exact solutions for the behavior of magnetic molecules. Likewise Costa Soukoulis uses analytical and numerical techniques in his studies of disordered systems (e.g., random lasers). Jörg Schmalian uses many body techniques to study correlated electron systems (e.g., the pseudogap in high temperature superconductors) and is studying defect induced inhomogeneities and glassiness in such systems.

With the amazing increases in computational power and with the many successful applications, the field of computational materials science has grown enormously. Unfortunately it is not yet a fully developed community. Two years ago the Computational Materials Sciences Network (CMSN) was set up to support teams of researchers to come together across interdisciplinary and inter-institutional lines and work together on large and important problems. Ames is the lead lab in coordinating CMSN. So far, 5 separate projects have been funded with about 15 to 20 scientists working in each.

VII. Quasicrystals

Discovered as metastable phases in 1982, quasicrystals (qcs) are *atomically-ordered metallic alloys* that *lack periodicity*. They exhibit classically-forbidden symmetries, often five-fold. Thermodynamically-stable phases began to emerge in the late 1980s, at which time scientists realized that qcs possess unusual combinations of physical properties— apparently related to their remarkable atomic structure. Ames' history in this field began when Alan Goldman—who had been working on qcs almost since their discovery—joined our staff in the late 1980's. He and several scientists in Metallurgy and Ceramics (M&C) began a collaboration that paved the way for a "New Initiative" in Surface and Interface Properties, awarded by BES in FY97. This provided seed funding for research in fundamental surface and interface science, tribology, and coating development. Since then, interests in Ames have blossomed to include sample growth, bulk structure determination, electronic stabilization, magnetism, nucleation of qcs in solid and liquid phases, and polymer-qc composites. Today, Ames Laboratory is recognized internationally as a leading center of research in qcs and related complex intermetallics. This recognition is demonstrated, for instance, by our participation in *many* international workshops and conferences (often as organizers or invited speakers), and by *frequent* requests for collaboration. The leadership we enjoy is due to the productive, closely-knit interdisciplinarity within the project. Although the work is coordinated through Materials Chemistry (Pat Thiel), scientists from all three Materials Sciences Programs are equally important, and communicate frequently. Select highlights follow.

Where are the atoms? This question has nagged the field since its inception, because qcs do not lend themselves to conventional techniques for structure determination. John Corbett has recently developed a promising method for direct structure determination using x-ray diffraction. This approach, combined with transmission electron microscopy by Matt Kramer (M&C), has yielded a model for a previously-unsolved icosahedral alloy. Another approach, taken by Gordie Miller, is to infer the structure of qc phases from closely-related crystalline phases. Such understanding of structure may lead to understanding of electronic stabilization, and thence to predictive capability.

Bulk structure determination forms a springboard for surface structure determination. Several years of work have led us to conclude that, for the Al-rich icosahedral alloys, the clean surface can be prepared as a relatively flat structure in which the atomic arrangement is indistinguishable from a bulk-terminated structure. We have also characterized the oxidized surface that forms in air. These studies form a basis for studies of tribology, wetting, and film growth. We have recently found evidence that the low coefficient of friction of quasicrystals is, at least in part, an intrinsic property of the clean, bulk-terminated surface. This work is done by Cynthia Jenks and Pat Thiel, with numerous on-site collaborations (e.g., Sordelet, Kramer, Goldman) and major contributions from two off-site collaborators: Michel Van Hove (structure determination, LBNL), and Andrew Gellman (tribology, Carnegie Mellon University).

For many types of studies, including surface studies, large defect-free single grains are necessary, but rarely available. We have achieved several breakthroughs, including implementation of flux growth of quasicrystals, which was a very significant advance in the field. Our samples, both flux- and Bridgeman-grown, are in demand worldwide. This work is carried out by Tom Lograsso and Amy Ross (M&C), and Paul Canfield, with important feedback from Cynthia Jenks, an on-site user of the samples.

VIII. Polymers and Organic Materials

The area of polymers has expanded recently in the Materials Chemistry program. For many years, Tom Barton worked in the area of silicon- and germanium-containing polymers, striving (in a bottom-up-type approach) to understand the fundamental chemical reaction mechanisms, to find new synthetic routes, to optimize yields, and to characterize and optimize properties of his new materials. He achieved significant successes, but was the only ‘polymer chemist’ in the Laboratory. In recent years, Materials Chemistry has added Valerie Sheares, Klaus Schmidt-Rohr, and Surya Mallapragada. [This group of young scientists has received an impressive set of awards over the last 3-4 years, ranging from “Young Investigator” awards (Dow, 3M, NSF, Sloan, Beckman), to the Dillon Prize for Polymer Physics from the American Physical Society.] Their unifying goal is to predict and tailor behaviors of polymers. To this end, they bring together complementary areas of polymer expertise: synthesis (Sheares), characterization (Schmidt-Rohr), and bio-engineering (Mallapragada).

One theme emerging among this group is biomaterials. Valerie Sheares has discovered that quasicrystals are a very promising wear-resistant additive for polymers. (This is mentioned again in Section VII, “Quasicrystals.”) Sheares and Mallapragada are collaborating now on developing these polymer-quasicrystal composites as biomaterials, specifically as bone substitutes in hip replacements.

Biomaterials are also the driving force in the development of environmentally-responsive polymers, particularly polymers that undergo swelling/syneresis in response to environmental conditions such as changes in pH (Mallapragada). Such polymers could be very useful for the treatment of diseases such as insulin-dependent diabetes. Ideally, the gels would swell and release insulin whenever glucose levels in the body increase and the pH decreases. This project will take advantage of Schmidt-Rohr’s expertise in NMR (below) to characterize the crucial micellization and gelation dynamics of the polymers.

Because polymers are such complex materials, it is not easy to measure or understand their structure and dynamics on a molecular scale. Neutron scattering is one technique often used for this purpose; another is solid-state nuclear magnetic resonance (NMR). Our emphasis is on the latter. Schmidt-Rohr uses NMR to study heterogeneous polymer systems, in particular nanocomposites of organic polymers and inorganic species. His recent NMR technique development has produced several methods that are useful in such studies.

Understanding the basic mechanisms governing the radiation properties of organic materials and the conducting processes in molecular and polymeric films is a prime topic for Joseph Shinar. Such fundamental studies underpin the successful pursuit of flexible organic transistors and efficient light emitting devices. J. Shinar and T. Barton work together closely in developing and testing new opt-electronic polymers and oligomers.

Highlights

Antropov

Electronic Structure and Magnetism (theory)

Description: The principal objective of this program is the study of magnetism and electronic structure of metals, their alloys and compounds using modern methods of computational physics (density functional, GW and so on). The phenomena studied in ab-initio calculations are electronic structure transformations under influence of different perturbations: pressure, defects, structural and magnetic disorder. After electronic structure is identified, the parameters of magnetic interactions are calculated using ab-initio methods to be used in model spin simulations(Monte-Carlo, Nose-Hoover and so on). These parameters are useful to describe systems with many atoms (micron scale). Properties studied on this length scale are domain wall structures, their pinning and coercive force.

Highlights:

- *Electronic structure and superconductivity in MgB2.* Our first electronic structure calculations described the coexistence of covalent and metallic bonding in this compound, similar with intercalated graphite, and predicted electron-phonon coupling as the main mechanism for superconductivity
- *Spin dynamics.* We continue to improve algorithms of spin dynamical simulations in molecules and solids. Numerous calculations of exchange coupling and anisotropy parameters have been performed using the multiple scattering formalism (CoPt, FePt, SmCo5, Nd-Fe-B, MnAl and others). These parameters are being used in the model simulations. Giant anisotropy in the R-Co5 system has been found in local density approximation. For the first time a GW-like approach has been successfully applied to describe transition metal magnetism (Fe, Ni, Gd).
- *Domain walls dynamics.* We were the first to propose a working scheme to connect different scales in magnetism for real systems: atomistic (10-100 atoms) and model (up to millions) spin calculations. This scheme has been applied to study domain wall dynamics in CoPt and related compounds. The structure of the domain wall and possible sources of pinning have been identified.
- *Highly responsive materials (Gd-Si-Ge)*. We determined the character of chemical bonding and exchange coupling parameters in these materials.

Impact: Our results have been used in complimentary theoretical and experimental research. Obtained parameters of Heisenberg model are being used by other groups in model spin simulations. Developed formalism for spin dynamics and exchange parameters calculations are being used in many groups (ORNL, SNL, NRL and others)

Interactions:
U.S.National Laboratories: Dept. of Metallurgy and Ceramics, Materials Chemistry (Ames Lab); Sandia Livermore Nat. Lab, CA; Naval Research Lab, DC; Oak Ridge Nat. Lab, TN.
Non-U.S. Laboratories: Kurchatov Institute, Moscow,Russia; Lebedev Institute, Moscow, Russia; Insitute of Physics of Metals, Kiev (Ukraine)

Personnel: V. Antropov (group leader); K. Belashchenko, G. Samolyuk (postdocs), N. Zein, V.Antonov (visiting scientists), E.Riabinkin (student)

Recognition:
- 2 invited review articles in last 2 years
- 8 invited talks at national and international conferences in last 2 years

Biswas

Photonic Band Gaps

Description: The principal objective is to design, model and fabricate novel periodic structures resulting in photonic crystals that can manipulate and control light in unique ways. Novel procedures are being developed to synthesize photonic crystals operating at infrared and optical wavelengths. The interaction of electromagnetic waves with these photonic crystals is simulated with band calculations, transfer matrix methods and finite difference techniques. Simulations are critical in interpreting the results of measurements and in designing new structures with desired electromagnetic behavior. A close collaboration with Sandia National Laboratories is being pursued where simulations at Ames are being used to fabricate and model new photonic crystals at Sandia.

Highlights:

- *Structure of thin film photonic crystals*: Theoretical calculations have been combined with optical and X-ray measurements to determine, for the first time, the structure of self-assembled thin film photonic crystals. An unusually large compression along the growth direction was found that now explains many previously conflicting pieces of experimental data.
- *Exceptionally directional sources*: Exceptional directional sources with beam widths less than 12° and high frequency selectivity, were designed and fabricated by utilizing the resonant cavities formed from photonic band gap crystals.
- *Collaboration with Sandia on infrared photonic crystals*: Theoretical calculations at Ames have led to the fabrication of a new metallic photonic crystal and a ultra-small single-mode photonic crystal cavity at Sandia, operating at infrared wavelengths.
- *Infrared sources*: In collaboration with Ion Optics we have designed patterned surfaces that act as extremely narrow band thermal emitters at infrared wavelengths by utilizing the sharp surface plasmon resonances on the periodic surface textures.

Impact: The Ames Lab 3-dimensional photonic crystal design has been employed by laboratories throughout the world. Our research results have motivated complementary theoretical and experimental investigations internationally. Since the initial simulation and demonstration of an actual dielectric structure exhibiting photonic band gap properties there are over 500 PBG groups worldwide with web pages: (http://www.neci.nj.com/homepages/vlasov/pbgroups.html)

Interactions:
Ames Laboratory: A. Goldman, J. Shinar
U.S. National Laboratories: Sandia National Laboratory, Advanced Photon Source (Argonne National Lab).
Iowa State University: Dept. of Electrical and Computer Engineering, Microelectronics Research Center
U.S. Universities: University of Utah, Northwestern University
Non-U.S. Universities: Bilkent University (Turkey); U. of Amsterdam, U. of Crete
Industrial: Agilent Technologies, Ion-Optics Inc, Honeywell

Personnel: K.M. Ho, C.M. Soukoulis, R. Biswas (theory) (group leaders), K. Constant, G. Tuttle, W. Leung (experiment); I. El-Kady, S. Foteinopoulou, H. Kang, K. Sutherland, C.-H. Kim, Y.-S. Kim (graduate students)

Recognition:

- Energy 100 award with rank 24; Science 100 award of the DOE.
- Organized NATO Advanced Research Institute on PBG's June 2000 (C. M. Soukoulis, director)
- "*Photonic Frenzy,*" Ames Laboratory Inquiry magazine, Fall 1999
- 6 Invited talks at national and international conferences last year

Borsa

Nuclear Magnetic Resonance in New Materials and Phases

Description: The aim of this program is the study of the microscopic static and dynamic properties of new materials and phases particularly regarding their magnetic properties. The systems recently investigated include the high Tc copper oxide superconductors as well as the newly discovered MgB_2 superconductor, the glassy superionic conductors and the novel molecular nanomagnets. Nuclear Magnetic and Quadrupole Resonance is a technique that yields local microscopic properties and is complementary to other techniques for the investigation of materials. Thus the mode of operation involves a broad cooperation with other groups in Ames Lab both experimental and theoretical.

Highlights:

- *Glassy spin freezing and stripes in cuprate superconductors*. Our ^{139}La and ^{65}Cu NQR and NMR studies, in connection with magnetic susceptibility data, has allowed the determination of the phase diagram in underdoped lanthanum cuprate system and it has provided forceful early although indirect evidence for the microsegregation of holes in “rivers of charge” (stripes)and for the coexistence of a cluster spin glass phase and a superconducting phase.
- *Magnetic structure of the ground state in molecular nanomagnets Mn12 and Fe8.* From the proton and ^{55}Mn NMR spectra at low temperature we have obtained information about the local orientation of the magnetic ions in the high spin (S=10)quantum ground state.
- *Spin dynamics in molecular nanomagnets.* We have shown that measurements of nuclear spin-lattice relaxation provide information about the spin dynamics in a large number of molecular nanomagnets both with antiferromagnetic singlet ground states (Fe10,Fe6, Cu8)and with magnetic ground states (Cu6, Mn12, Fe8).
- *Quantum tunneling of the magnetization in molecular nanomagnets.* We have demonstrated a novel method to monitor the low temperature relaxation and the quantum tunneling of the magnetization by using the local NMR probe.
- *Speciation of mobile ions in glassy fast ionic conductors.* We have shown for the first time the existence of different channels for the motion of ions in a silver borate mixed fast ionic conductor.

Impact: Our NMR research in molecular magnets has triggered the wide effort both in theory and experiments going on presently in Ames Lab. It has motivated Prof. D’Alessandro in the Department of Mathematics and Prof. Salapaka in Electrical Engineering (both ISU) to investigate quantum control of molecular magnets. Finally, it has stimulated the interest of four iInternational laboratories, which, as a consequence, have undertaken NMR research in molecular magnets (C. Berthier in Grenoble; K. Kumagai in Hokkaido, Japan; T. Goto in Kyoto, Japan; B. J. Suh in Seoul, Korea)

Interactions:
Iowa State University: Ames Laboratory (D. C.Johnston, P. Canfield, M. Luban, P. Kogerler, S. Dobrovitsky, L. Miller); Department of Material Science and Engineering (S. Martin); Department of Mathematics (D. D’Alessandro); Department of Electric Engeneering (M. V. Salapaka)
Non-U.S. Universities: U.of Pavia (A. Rigamonti, A. Lascialfari) and U. of Florence (D. Gatteschi, A.Cornia) , Italy; U. of Grenoble, France (M. H. Julien); U. of Hokkaido (Y.Furukawa) and U. of Kyoto (T. Goto), Japan; Ecole Politechnique de Lausanne, Switzerland (C. Dimitropoulos); U. of Trondheim, Norway (I. Svare)

Personnel: F. Borsa (group leader); R. Vincent (visiting scientist); J. K. Jung (postdoc), D. Proscissi, B. Meyer, Seung-Ho Baek (students)

Recognition:
- 10 invited talks at international universities and international conferences in the last 2 years
- 1 invited review paper in 1998

Canfield

Design, Discovery, Growth, and Characterization of Novel Materials

Description: Design, discovery, growth and characterization of new and exotic materials with electronic and/or magnetic phase transitions is the primary focus of this group. Compounds are usually prepared in single crystal form, so the measurement of the anisotropic, temperature and applied magnetic field dependent magnetization, specific heat and electrical resistivity are our primary characterization measurements. Currently, the majority of the research effort is directed to the study of the interaction between 4-f electrons and conduction electrons. This interaction can be studied in several different classes of systems: magnetic superconductors, mixed valent and heavy fermion compounds, quasicrystals and intermetallic compounds with moment bearing rare earth ions. In each case the interplay between the conduction electrons and 4-f moments manifests itself in a different manner. Once full magnetization, specific heat, and resistivity measurements have been completed, a wide range of collaborations are possible. These include ISU/Ames Lab collaborations, as well as collaborations with a large number of national and international research groups. These collaborators are experimentalists with specific skills who are interested in well characterized samples that manifest interesting physical properties. In addition, the high purity and single crystal form makes collaborations particularly desirable and make it possible to rapidly gather large amounts of data on "hot" (e.g., recent work on MgB2) materials and have helped us to assume an international presence in several areas of condensed matter research.

Highlights:

- World leadership in research on RNi2B2C magnetic superconductors.
  -interaction between magnetic and superconducting ground states
  -non-local effects and structure of Flux Line Lattice
  -phonon softening and phonon mode coupling
- MgB2 work—promptly delveloped technique to produce high purity, very low resistivity powders and wires of MgB2. Over three weeks we published three PRL's on mechanism, properties, and wires of MgB2.
- Solution growth and characterization of single grains of RMgZn and other intermeatllic quasicrystals.
  -First single grains of RMgZn and characterization of spinglass state.
  -Growth and characterization of large grains of AlPdMn, AlNiCo, and AlPdRe quasicrystals.
- Growth of wide variety of intermetallic compounds in single crystal form for internal research projects as well as external collaborations.

Impact: By producing high purity samples (single crystal, single grain, powder) and performing detailed measurements and analysis this group has helped create several active relms of research. Three systems RNi2B2C, RMgZn, MgB2 are particularly good examples. By providing samples to collaborators on a global basis this group helps to support and influences research at an international level. Basically we publish our own good work and then feed the world.

Interactions:
As a result of being able to produce some of the worlds best samples of specific intermetallic materials, this group collaborates extensively with other groups in Ames Lab, other DOE Labs, other research groups in U. S. Universities and other research groups internationally.

Personnel: P. C. Canfield (group leader); S. L. Bud'ko (Scientist); I. R. Fisher (now at Stanford); C. Petrovic (Post-Doc); G. Lapertot and C. E. Cunnigham (Visiting Researchers); Graduate Students: K. Myers, T. Wiener, X. Miao, C. Gao; Undergraduate Students: N. Anderson, N. Kelso, P. Mott, Angela Hvidvid, Jason Schissel

Recognition:

- 1998 – present: 30 invited talks
- 1998 – present: 11 PRL, 30 PRB
- Recent MgB2 work discussed in *Science, Nature, Physics Today, Physics World, New York Times, Washington Post, A. P.,* and several European and Japanese newspapers.

Clem

Superconductivity—Theory

Description: The objective of this program is to advance theoretical understanding of the behavior of superconductors in magnetic fields, by studying critical currents and fields, ac losses, anisotropy, and the structure of vortices and vortex lattices. These are essential components of theory needed for interpreting experiments and designing applications of superconductivity. Choices of topics are usually guided by experiment.

Highlights:
- Pioneered in developing theory explaining that ceramic samples of the high-temperature superconductors can be characterized as granular materials consisting of strongly superconducting grains, which are Josephson-coupled via weak links.
- Developed a new description of vortices in the layered superconductors in terms of intralayer two-dimensional *pancake vortices* (a name we coined) connected from layer to layer by interlayer Josephson strings.
- Developed a theory predicting that to reduce noise from trapped vortices in SQUIDs cooled in the earth's magnetic field, the width of all superconducting lines should be less than about 5 mm.
- Developed methods for calculating the magnetic fields generated by vortices in superconducting thin films, thin-film annular rings, and films with grain-boundary junctions, including superconductors with d-wave symmetry.
- Developed theory for ac losses in superconducting power transmission lines.
- Developed theory for the effect of *geometrical barriers* and *field-dependent Bean-Livingston barriers* on the penetration of magnetic flux into superconducting strips and disks.
- Served as Science Editor and regular contributor of the "Nota Bene" section of *High-T_c Update*, which for 13 years highlighted interesting preprints on high-temperature superconductivity. Serving as Science Consultant to the *High-T_c Update* web site, which is highlighting MgB_2.

Impact:
- Recognized in the community as one of the leaders in the field of magnetic properties of type-II superconductors.
- The *pancake vortex* is now an essential part of the vocabulary of high-temperature superconductivity.
- Citations to Clem papers: 525 in 1998, 487 in 1999, and 493 in 2000.
- Our prediction that SQUID noise can be sharply reduced by using narrow superconducting lines was confirmed in experiments in the group of J. Clarke (LBNL), and this concept is currently being used by SQUID researchers worldwide to design low-noise SQUIDs operating at 77 K.
- *High-T_c Update* had an important impact on the development of high-temperature superconductivity, identifying key problems, providing timely information, and fostering international collaborations.
- Co-organizer of International Workshop on AC Losses in High-Temperature Superconductors, Palo Alto, California, April 8-9, 1999, and 9th International Workshop on Critical Currents, Madison, Wisconsin, July 18-21, 1999. Served on Program Committee or International Advisory Committee of nine additional international conferences in 1998-2001.

I**nteractions**:
I<u>ntralaboratory</u>: With the experimental groups of D. K. Finnemore, D. C. Johnston, and P. C. Canfield. Theoretical interactions are with V. G. Kogan.
<u>Outside</u>: Pirelli Cable Company, IBM, Los Alamos National Laboratory, Electrotechnical Laboratory, MPI-Stuttgart, University of Bath, University of Nottingham, Universite' Paris-Sud, Stanford University, Tel Aviv University, and University of Tokyo.

Personnel: J. R. Clem (group leader); Y. Mawatari and A. A. Babaei Brojeny (visiting scientists).

Recognition:
- Fellow of the American Physical Society.
- APS Division of Condensed Matter Physics Executive Committee, 1992-96; Chairman, 1994-95.
- Department of Energy 1988 Annual Award for Sustained Outstanding Research in Solid State Physics.
- Special award from the 1990 Applied Superconductivity Conference, Inc., for contributions to the superconductivity community via *High-T_c Update*.
- One patent.
- 30 invited talks 1998-2000.
- Chair.

Finnemore

Superconductivity

Description: The principal objective of this program is to study the fundamental interactions that control superconducting correlations in materials. We want to understand the fundamentals well enough to be able to enhance the performance characteristics and to create new materials with superior electrical and mechanical properties. Specific problems of current interest are supercurrent transport at grain boundaries, and flux pinning at defects in superconductors. Of primary interest are the factors that control the transition temperature, the superconducting order parameter, the characteristic magnetic fields (H_c, H_{c1}, & H_{c2}), the characteristic lengths (λ,ξ), and the strength of the forces that pin superconducting vortices at defects.

Highlights:

- For the new 40 K superconductor, MgB_2, the fundamental values of the upper critical field, H_{c2}, the thermodynamic critical field, H_{c2}, the penetration depth, λ, and the coherence distance, ξ, have been determined and found to be consistent with the properties of classical superconductors.
- Critical current densities in MgB_2 were were shown to be larger than 20,000 A/cm^2 at 20 K and 1 Tesla thus opening a new regime of performance for classical superconductors. Supercurrent transmission through grain boundaries is very easy, indicating that this material will need no grain orientation to get high performance.
- Boron fiber that is available in kilometer lengths at very reasonable cost was shown to produce very high performance MgB_2 wire when flashed with Mg vapor at 950^o C.
- The thermodynamic critical field of underdoped $La_{2-x}Sr_xCuO_4$ was measured for x-values over the full range where superconductivity occurs and the free energy was shown to scale with the transition temperature even when the carrier concentration is reduced by a factor of ten. The Cooper pairs become very two-dimensional with underdoping, but the free energy relations are similar to classical superconductors.
- Vortex fluctuations at temperatures close to T_c in $La_{2-x}Sr_xCuO_4$ were shown to change from 2D behavior at low field to 3D behavior as the field approaches H_{c2}. As the c-axis coherence distance becomes larger than the copper-oxide plane spacing, the 2D pancake vortices become real line vortices and 3D behavior is observed.

Impact: The ease of supercurrent transmission through grain boundaries in MgB_2 and the ease of transforming commercial boron fibers into high quality material opens the door to the development of a low-cost conductor for superconducting instruments that operate at 20 K. We developed the process to create MgB_2 wire by exposing boron fiber to Mg vapor, a process now commonly used in Europe and Japan.

Interactions:
Industry: Textron Systems in Wilmington, MA, has been a good working partner in the development of MgB_2 conductors.
<u>Local</u>: Canfield, Bud'ko, Kogan, Kramer, and McCallum are important collaborators in the MgB_2 project.

Personnel: D. K. Finnemore (group leader), Y. Huh (student), and J. E. Ostenson (scientist)

Recognition:

- fellow of APS
- 3 invited talks
- 1 invited review article

Goldman

X-ray Scattering

Description: Our group uses the techniques of resonant and nonresonant x-ray scattering as a probe of magnetic ordering in solids and uses both x-ray and neutron scattering techniques to study magnetically ordered systems as well as other novel compounds. We continue to study the magnetic structures of crystals with the $ThCr_2Si_2$ structure including both the RNi_2Ge_2 and RCu_2Ge_2 families. These materials yield a wide variety of magnetic structures at low temperature in both zero applied magnetic field and in external fields. This behavior arises from the closely similar energy scales of the RKKY interaction and CEF levels. The ability to tune, or select the magnetic structure by a simple substitution of the rare earth element makes these compounds ideal for detailed studies of magnetic interactions.

Highlights:

A determination of the magnetic structure of $GdAgSb_2$ by neutron scattering techniques is made difficult due to the strong neutron absorption of naturally occurring Gd. Using resonant x-ray scattering, we found that the antiferromagntic order below T_N corresponds to a magnetic wavevector of (1/2 0 0), with the moment direction along [0 1 0]. Interestingly, the measurements were made using the quadrupole, rather than dipole, resonance at the Gd L_{III} absorption edge. This feature is generally quite weak, but the measurements were made possible by the high brilliance of the undulator line at the APS.

We have reinvestigated the magnetic ordering in this compound. Neutron scattering studies, which did not have sufficient Q-resolution to observe the distortion, found that the antiferromagnetic structure corresponded to a longitudinally polarized spin wave along the in-plane [1 0 0] directions. In the orthorhombic phase, however, the in-plane degeneracy is broken allowing a unique determination of the magnetic wavevector. Through high-resolution resonant scattering measurements we determined that the magnetic wavevector and moment direction were, in fact, found along the longer basal plane axis. Theoretical calculations of the change in Fermi-surface nesting in the orthorhombic phase found good agreement with these observations.

Previous magnetic neutron scattering measurements of $TbCu_2Ge_2$ found a simple antiferromagnetic structure below T_N=13K with the magnetic wavevector (1/2 0 1/2), and the moment direction along [1 1 0]. Recent magnetic susceptibility measurements, however, found evidence of a second transition at T_t=9.3K. Resonant and nonresonant magnetic scattering at the Tb L_{III} absorption edge of a small single crystal sample found that this transition corresponds to a reorientation of the magnetic moment. Below T_t, the moment is along the [1 1 0] direction. As the temperature is increased and approaches T_t, the moment rotates towards the [1 0 0] direction and locks in for temperatures between T_t and T_N.

Impact: Using the MUCAT undulator line at the Advanced Photon Source, our group has made significant progress in developing magnetic x-ray scattering as a technique for the determination and refinement of antiferromagnetic structures.

Interactions:

<u>Ames Laboratory:</u> Canfield, Stassis, Harmon, Antropov, Kramer, McCallum, Ho, Thiel, Sordelet, Vaknin
<u>U.S. Universities</u>: University of Missouri, SUNY Stony Brook, Georgia Tech, Kent State, Michigan State, University of Wisconsin, Washington University.
<u>U.S. National Laboratories:</u> Argonne, Brookhaven
<u>Non-U.S. Laboratories</u>: F.Z. Juelich

Personnel: A.I. Goldman (group leader), D. Wermeille, A. Letoublon (Post docs), C.Y. Song, W. Good, J. W. Kim (graduate student)

Recognition:

- 1 Fellow of the APS
- 3 DOE Materials Science Awards since 1995
- 3 invited talks at national and international conferences

Harmon

Computational Materials Science

Description: Remarkable advances in computing power are providing opportunities to accurately simulate physical processes that are key for solving outstanding problems in science. The evaluation of the electronic structure of complex materials is a starting point for understanding the magnetic, conducting, optical, and structural properties of systems as a function of temperature, pressure, or applied fields. Many times we work with experimental groups (frequently borrowing their graduate students) to analyze and often predict the detailed and sometimes unusual behavior exhibited by modern complex materials. In the last few years time has been spent in helping coordinate the "computational materials science community". The motivation for this comes from the growth and success of the area, and also from the unnecessary duplication of effort in algorithm development and the need for professional level programming for modern parallel supercomputing applications.

Highlights:

- *Magneto-optics:* 1) Analytically derived the result that a 90 degree Kerr rotation is impossible for homogenous materials with absorption. This led to explanations for the observed 90 degree Kerr rotation in CeSb based on thin oxide layers. 2) In a series of papers the MO spectra were calculated for a range of magnetic materials. We demonstrated that in systems where correlated electrons (mostly 4f's) the LDA+U approach gives superior agreement with experiment. In fact the MO spectra provide an excellent measure of the interactions of correlated states with itinerant electron states.
- *Magnetic Molecules*: Realistic modeling of the magnetic tunneling and relaxation processes in several magnetic molecules (Mn_{12} and V_{15}) are based on analysis of numerous experimental results. The Hamiltonian which includes interactions among the many magnetic ions (rather than the simplified single spin model) was shown to be needed for consistent quantitative agreement with the data. Long quantum coherence times were predicted for V_{15} , although there is some distance to quantum computing qubits.
- *Spin Dynamics:* First principles SD approach is used to provide data for model Hamiltonian SD for meso-scale systems containing thousands of atoms. In turn, we are developing coarse graining methods to tie the meso-scale to micro-magnetic algorithms (see CMSN).
- *CMSN:* The "Computational Materials Science Network" was established to promote interdisciplinary and inter-laboratory collaboration on important problems requiring such teams. We are involved in the CMSN project on magnetic materials.
- *Misc.* Calculations in collaborations with various groups in Met. and Cer. and CMP have or are being done ($Gd_5Si_2Ge_2$, $AlMgB_{14}$, Ni_2MnGa, RNi_2Ge_2, etc.), with issues of Fermi surface nesting, magnetic coupling, bonding, etc., explored.

Impact: First principles "Spin Dynamics" has been adapted by several groups in the US (eg ORNL, U of Texas) and in Europe (eg Vienna, Stockholm). CMSN is successfully operating with five separate projects. Ran an Advanced Study Institute in Santiago (Jan. 2001) on CMS.

Interactions: Ames Lab: Antropov, Goldman, Lynch, Canfield, Borsa, Stassis, Gschneidner.
U.S. National Laboratories: ORNL, Argonne, Sandia Livermore, LANL
Non-U.S. Institutes: ESRF, Grenoble; Institute of Metals, Ekaterinburg; Lebedev Inst., Moscow.

Personnel: Staff: B.N. Harmon (group leader), V. Dobrovitsky (scientist); Y.B. Lee, M. El-Saqer;(graduate students); M. Katsnelson (Ekaterinburg), Y. Uspenski (Lebedev Inst.), V. Antonov (Inst. of Metals, Kiev), J.Y. Rhee (Hoesio U., Korea) (visiting scientists).

Recognition:
- Fellow of the APS, on Executive Board of DCMP of the APS until 2003.
- 12 invited talks in the last two years.
- Recent review panels: Advanced Photon Source (4yr), BNL, SNL, NRL, LANL (CMP programs)
- Co-recipient of 1996 DOE/MS award for "sustained research"

Ho

Condensed Matter Physics (theory)

Description**:**
Atomistic modeling and computer simulations of fundamental interactions of solids and solid surfaces
Theory and fabrication of photonic band gap materials

Highlights:

Tight-binding molecular dynamics. Our group pioneered development of tight-binding potential schemes for molecular dynamics simulation of various condensed matter systems.("Sustained Outstanding Research award in Solid State Physics" DOE Materials Science Division, C. Z. Wang and K. M. Ho 1996). Recent development of potentials transferable to semiconductor surfaces allow us to study a number of challenging problems using a combination of tight-binding studies and first principles LDA calculations such as adatom and addimer diffusion on semiconductor surfaces, step structures, as wellas non-thermal graphitization of diamond surfaces under femtosecond laser pulses. On going studies include magic clusters on silicon surfaces, behavior of adatoms and adatom clusters near steps, adatom and addimers diffusion and incorporation in homo and heteroepitaxial growth, quantum size effects in metal thin film growth on semiconductor surfaces, grain boundary structures. ~19 papers (10/98 to present) in this area including 5 papers in PRL. Collaborations with Hupalo and Tringides (Ames), Feng Liu and Lagally (Madison, Wisconsin).

LDA calculations and class molecular dynamics. Our group has expertise in first principles density functional calculations in applications to surface and bulk condensed matter systems and classical molecular dynamics calculations. Areas where we have made significant contributions are: metal surface reconstruction, metal overlayers on semiconductor surfaces, frozen-phonon calculations for transition metals, martensitic transformations. Ongoing work in classical molecular dynamics focuses on surface energy anisotropy at the solid/nmelt interface. Collaborations with J. R. Morris, Trivedi, Napolitano, Kramer, Akinc (Ames), C. L. Fu, M. H. Yoo (ORNL), Elsasser, Faehnle (Stuttgart).

Genetic algorithm. Pioneered the application of genetic algorithms to structural optimization problems in condensed matter physics. Recent applications to semiconductor clusters in collaboration with Shvartsburg and Jarrold (Northwestern U) establish the structural motif for prolate Si, Ge and Sn clusters in the size range 10-20. ~9 papers (10/98 to present) including 3 in PRL.

Photonic band gap materials. Our role in the development of photonic band gap materials in theory initially and later on in experiment was recognized in the Energy 100 and Science 100 Awards from DOE. E. Ozbay (now at Bilkent U, Turkey) received the Adolph Lomb Medal from the American Optical Society for experimental work done while he was a member of our group in the area of photonic band gap material. ~15 papers (10/98 to present) in this area and 2 review articles. 2 invention disclosures (10/98 to present). Overall: 2 patents issued, 2 patents in prosecution. Collaborations with K. Constant (Materials Science, ISU), G. Tuttle (EE, ISU), R. Biswas and C. Soukoulis (Ames), Shawn Lin (Sandia), DP/BES nanoscience network with SNL, ORNL, LANL, LLNL, ANL, INEEL, industrial collaborations include Ion Optics (Massachusetts), Agilent Labs (Palo Alto).

Recognition:

- Fellow of APS
- ~700 citations per year
- DOE-BES nanoscience/nanotechnology group
- Board of editors, *Surface Review and Letters*
- External examiner, Physics Dept. Univ. of Hong Kong.

Johnston

New Materials and Phases

Description: The principal objective of this program is to develop new and improved solid materials that show novel or enhanced physical properties. Our physical property studies emphasize the magnetic properties of the materials, although we also carry out studies of the thermal and electronic transport properties. We have developed strong collaborations with many groups within Ames Laboratory and outside to leverage our research. Interfacing physics theory and experiment is an important component of our work.

Program Highlights:

- *Layered cuprate oxyhalides $M_2CuO_2Cl_2$ (M = Ca, Sr).* Our group was the first to recognize the importance of this previously known but unstudied class of layered cuprate materials. We developed procedures for growing single crystals. Our magnetic characterizations of them, with F. Borsa's group and D. Vaknin, helped to establish the intrinsic magnetic properties of undistorted CuO_2 layers in the layered cuprates. Our crystals have been in strong demand by other groups. These materials are now regarded by the CMP community as the best realizations in nature of the spin 1/2 square lattice Heisenberg antiferromagnet and as a prototype for the undoped single-layer high-T_c cuprate superconductor parent compound.
- *Stripe physics*. We discovered a scaling of magnetic susceptibility versus temperature $\chi(T)$ data for $La_{2-x}Sr_xCuO_4$ which indicated that the doped holes segregate into walls separating undoped, locally antiferromagnetically ordered, domains. Subsequent work over many years with F. Borsa's group and others supported this picture. Our studies strongly contributed to the development of "stripe physics", an important current paradigm in the study of cuprate high-T_c superconductors, nickelates, and manganites.
- *LiV_2O_4*. From our intensive effort to purify the material and a variety of physical property measurements with F. Borsa's group and others at U.C. San Diego, Columbia U. and Argonne Nat. Lab., the spinel structure compound LiV_2O_4 was found to be the first *d*-electron compound to exhibit heavy fermion behavior at low temperatures. This material contains no *f*-electrons and thus violates conventional wisdom about the nature of the materials in which heavy fermion behavior is expected to occur.
- *Spin ladders*. With experimentalists and theorists in Europe and Japan, we carried out extensive numerical studies of $\chi(T)$ of spin 1/2 Heisenberg two-leg ladders as well as measurements of $\chi(T)$ for all known inorganic (cuprate and vanadate) two-leg spin ladder compounds. From a comparison of the theoretical and experimental results, we found that an exchange interaction in addition to the usually assumed nearest-neighbor Heisenberg interaction is likely important to the magnetic properties of the materials, and we identified the so-called four-spin cyclic exchange interaction as a probable candidate.
- *Spin chains*. Numerical and experimental studies of $\chi(T)$ and specific heat of uniform and alternating-exchange spin 1/2 antiferromagnetic Heisenberg chains were carried out with colleagues in Europe and Japan. The numerical data for the uniform chain at low temperatures were found to be in excellent agreement with exact field theory predictions. We applied these results to model our experimental $\chi(T)$ data for the ambient (AP) and high (HP) pressure phases of $(VO)_2P_2O_7$. We inferred that the AP phase contains two distinct types of alternating-exchange chains, whereas the HP phase contains a single type.

Impact: Our results motivated a wide range of complementary experimental and theoretical studies. The impact of our research is reflected by the citation rate of our publications (~400/year) and the nature and number of the invited talks given (98 over the past decade).

Interactions:
Local: F. Borsa, P. Canfield, B. Harmon, T. Lograsso, M. Luban, D. Lynch, R. Modler, M. Porter, D. Vaknin
U.S. Universities: U. Illinois at Urbana-Champaign, Northwestern U., Columbia U., U.C. San Diego, U.C. Berkeley, Stanford U., Southeast Missouri State U., Colgate U.
Non-U.S. Universities: Queen's U., Hokaido U., U. Pavia, U. Tokyo, Kyoto U., U. Osnabruck, ETH Zürich, IFW Dresden
U.S. National Laboratories: Argonne, Los Alamos

Personnel: D. C. Johnston (group leader), L. L. Miller, C. A. Swenson (emeritus associate), J. M. Hill and M. Fitzpatrick (graduate students), M. Todd and G. Knoke (undergraduate students), W. C. Lee (one-year visitor)

Recognition:

- 2 Fellows of the American Physical Society (Johnston, Swenson)
- 5 invited talks at institutions and 6 at national and international conferences since 1 Oct. 1998
- 1 major recent invited review article (237 pages, 1997)

Kogan

Theory II: Superconductivity

Description: Develop a theoretical basis for understanding the effects of strong anisotropy on the magnetic properties of type-II superconductors in general and high-temperature superconductors in particular. Specific areas include:

- Fluctuations in strongly anisotropic layered superconductors
- Effects of nonlocal electrodynamics of superconductors on low temperature magnetization, vortex lattices, etc., in conventional and high-Tc superconductors.
- Theoretical basis for the scanning SQUID microscopy (SSM) for studying London and Josephson electrodynamics in s- and d-wave superconductors.

Highlights:

- Corrections to the London theory due to nonlocality of the current-field relation in superconductors were derived. This is a new theoretical tool for studying "vortex solids," an important part of vortex physics.
- Phase transitions in the vortex lattice structure were predicted and subsequently confirmed by extensive small angle neutron scattering and decoration experiments in superconducting tetragonal borocarbides.
- Models developed for interpretation of scanning SQUID microscopy data made this new experimental technique into a quantitative tool for studying superconductivity (extracting the interlayer penetration depth directly from images of the in-plane vortices in layered superconductors, clarifying the nature of the random telegraph noise observed with SSM in mesoscopic superconducting structures).
- Nonlocal Josephson electrodynamics for linear junctions in thin films has been developed for understanding properties of the grain boundary junctions, used extensively in studies of unconventional superconductivity.

Impact:

- Our work on superconducting anisotropies and in particular on torque magnetometry and fluctuations are well-known and used by the world community.
- Our work on nonlocal effects in superconductors developed into a new field of vortex research, which we are leading.

Interactions

- Nonlocal effects on flux-line lattices and macroscopic magnetic properties of borocarbides: P.Canfield, I. Fisher, S. Bud'ko (Ames)
- Neutron scattering from vortex lattices: P. Gammel and D. Bishop (Lucent Technologies), M.Eskildsen (Switzerland), D. McK. Paul (UK)
- Theory of vortex lattices in superconductors: P. Miranovich (Montenegro), A. Gurevich. (Madison)
- Josephson phenomena: R. Mints (Tel Aviv), J. Clem (Ames), Y. Mawatari (Tsukuba)
- SQUID microscopy: J. Kirtley, C. Tsuei (IBM), K. Moler (Stanford), J. Clem (Ames).

Personnel: V. Kogan (group leader)

Recognition

- Work is cited in Tinkham's textbook on superconductivity.
- Chapter on "Vortex lattice transitions" in a book on Superconducting State in Magnetic Field.
- Fellow of APS

Luban

Magnetism—Magnetic Molecules

Description: The principal objective of this program is to achieve a basic understanding of the magnetic properties of a class of newly synthesized materials referred to as magnetic molecules or molecular magnets. As a result of recent successes in polyoxomolybdate chemistry it is now possible to embed large symmetric arrays of paramagnetic ions (currently as many as 30) in diamagnetic host molecules. With the ability to control the placement of magnetic moments of diverse species within stable structures, entirely new magnetic phenomena are being observed. Some of these novel systems may even offer the prospect of useful applications. These systems thus present an ideal "laboratory" to explore in a controlled manner the bridge between microscopic, atomistic magnetism and cooperative phenomena encountered in bulk magnetic structures. Our investigations are performed in close collaboration with other theorists, synthesis chemists, and experimentalists, and includes providing theoretical input for synthesis, magnetization, specific heat, NMR, and inelastic neutron scattering studies performed by AL personnel and our external collaborators.

Highlights:

- Using a *classical* version of the Heisenberg model of interacting spins we have obtained excellent quantitative agreement with the measured (at AL and NHMFL) magnetic properties (temperatures down to 0.1 K and fields up to 60 Tesla) of the molecular magnet {Mo72Fe30}, the largest paramagnetic molecule synthesized to date, with 30 paramagnetic Fe ions (spins $s = 5/2$). At low temperatures the spins achieve a highly symmetric frustrated ordering.
- We have developed a *quantum* model of the molecular magnet {Mo72Fe30} which is applicable for the low temperature regime (< 2 K). The predicted dependence of the magnetization on magnetic field is in excellent agreement with the results of experiment (fields up to 60 Tesla). In our model the low-lying excitations form a sequence of "rotational bands" with a predicted energy gap of 0.74 meV separating the two lowest bands, a feature soon to be put to experimental test using inelastic neutron scattering and EPR techniques.
- Our theoretical results based on the quantum Heisenberg model successfully explain the magnetism and spin dynamics of the molecular magnet {Cr4} as determined from local susceptibility and proton NMR measurements
- Our theoretical treatment of the molecular magnet {V6}, utilizing AL susceptibility and inelastic neutron scattering data, has provided an unambiguous assignment for the exchange constants. A predicted level crossing at approximately 60 Tesla has been confirmed at NHMFL.
- We have provided a comprehensive theory of the magnetic properties of the molecular magnet {Ni4}. A sequence of plateaus, associated with energy level crossings, are predicted for the magnetic field dependence of the magnetization at low temperatures. First experiments at AL and NHMFL confirm this feature.

Impact: Our research findings, achieved in close collaboration with some of the leading investigators world-wide in this emerging field, have spurred new experimental and theoretical studies, and led to the synthesis of several new interesting species of molecular magnets meeting pre-determined requirements.

Interactions:

U.S. University: Naval Postgraduate School (theory)
Non-U.S. Universities: Univ. Bielelfeld (A. Müller *et al,* chemical synthesis), Univ. Hokkaido (NMR), Univ. Osnabrück (theory), Univ. Pavia (NMR).
U.S. National Laboratories: ANL (R.A. Klemm), NHMFL (LANL) (N. Harrison and A. Lacerda)
Non-U.S. Laboratory: Verkin Institute – Kharkov (theory)
Industrial: Philips Research (Hamburg), Telelogic (Bielefeld)
Local: F. Borsa, S. Bud'ko, P. Canfield, D.C. Johnston, L. Miller, R. Modler, C. Stassis, J. Zarestky

Personnel: M. Luban (Group Leader), P. Kögerler (Res. Assoc.), E. (Oniu) Morosan (student)

Recognition:

- 10 invited talks since 10-1-98, including two national and international conference talks
- 1 invited review article since 1-10-98

Lynch

Experimental Magneto-optics

Description: Magneto-optic Kerr effect (MOKE) spectra, Kerr angle and ellipticity, are measured in the 0.5 - 5-eV spectral range in fields up to 7 T. Sample temperatures range from 300 to 2 K. From such spectra and the dielectric function spectra (measured by ellipsometry) the diagonal and off-diagonal components of the optical conductivity are determined. The off-diagonal component, about 1% of the diagonal component, is a very sensitive test for calculated wave functions. Unfortunately, many interesting materials contain rare earths, for which modern computation methods are not reliable due to correlation effects in the 4f subshell, although La, Lu, and Gd may be treated well, the latter because the 4f states are all far from the Fermi level. Measurements of MOKE spectra in 4f and 5f systems, in fact, provide a good basis to compare with density-functional calculations using the local-density approximation (LDA). The failure of the LDA calculations is a direct consequence of correlated electron behavior and new methods such as the "LDA + U" approach are proving much more capable in describing MOKE in correlated-electron systems. Systems studied recently are RAl_3 and $R(TM)_2$, with R = rare earth, TM = 3d transition metal.

Highlights:

- Observation of a metamagnetic phase transition in an itinerant 4f system: $Ce(Fe_{1-x}Co_x)_2$
- Systematic study of role of rare-earth (R) 4f states in MOKE spectra of RAl_3 and $R(TM)_2$
- Determination that Fe_2VAl is not a heavy-fermion system, and that most of its unusual properties can be explained by site-interchange disorder

Impact: Demonstration that rare-earth 4f states are not directly involved in the MOKE spectra, but they are crucial because they polarize other bands. The correct theoretical treatment of the 4f states is needed to obtain agreement with MOKE data.

Interactions:
Non-U.S. Universities: Hoseo U., J.-Y. Rhee; Kon-Kuk U., K.-J. Kim (S. Korea)
Ames Lab: B. Harmon, V. Antropov, C, G, Olson, P. Canfield, K. Gschneidner, Jr.

Personnel: D. W. Lynch (group leader), Joon Mok Park, Jongik Park (graduate students). Recent Ph.D.: Ye Feng (student)

Recognition:

- Fellow of the APS
- Sincrotrone Trieste - Program Advisory Committee, 4 years
- Sincrotrone Trieste – Chair, Scientific Advisory Committee, 4 years
- Synchrotron Radiation Center—Users' Advisory Committee
- Several NSF panels (Science & Technology Centers, CAREER proposals, instrumentation grants)
- book: *Photoemission Studies of High-temperature Superconductors* by D. W. Lynch and C. G. Olson (Cambridge University Press 1999).

Modler

Complex Adaptive Materials in Extreme Environments

Description: The principal objective of this program is the study of fundamental microscopic interactions in condensed matter by means of transport and thermodynamic measurements, in the temperature range from 50mK to room temperature, magnetic fields up to 60Tesla, and pressures to 2GigaPascal. As we recently completed establishing our experimental capabilities, we are now able to *simultaneously* apply magnetic field (up to 11Tesla), high pressure and very-low temperature in order to *'tune'* complex/adaptive materials into a regime of critical instability. Since we also strive to *identify new complex/adaptive materials*, i.e. materials that show large responses to these parameters, we have developed a new generation of dilatometer allowing us to determine the effect of volume change on microscopic interactions. These new in-house experimental capabilities already proved to be extremely useful for our investigation of newly synthesized magnetic molecules at milliKelvin temperatures and in very-high magnetic fields.

Highlights:

- In the past year, we have completed our construction of a powerful experimental measurement system at Ames Laboratory that allows us to study materials under the *simultaneous influence* of external parameters high magnetic field (11T), very-high pressure (2Gpa) and very-low temperature (50mK). In the total range of simultaneous parameters, this system is one of very few and is targeted towards the investigation of the interplay between competing energy scales, such as magnetic and superconducting order parameters close to a quantum-critical instability.
- We have studied the magnetization of the so-far largest magnetic molecule $\{Mo_{72}Fe_{30}\}$ down to milliKelvin temperatures and ultra-high magnetic fields. In our experiments, we were able to observe (i) antiferromagnetic ordering of the geometrically frustrated system, (ii) its classical ground-state magnetization, (iii) the high-field saturation magnetization, as well as (iv) first traces of quantum behavior in this giant magnetic molecule (molar mass ~18649). Our experimental results are complemented by classical and quantum-mechanical calculations that are in excellent agreement with our experiments. Further investigation of the quantum behavior in $\{Mo_{72}Fe_{30}\}$ is under way, as well as magnetization studies of the novel magnetic molecules $\{Mo_{12}Ni_4\}$ and $\{V_6\}$.
- In our effort to investigate magnetovolume effects leading to the identification of novel complex/adaptive materials, we have developed a new generation of thermal expansion/magnetostriction capacitance cell manufactured from highest-purity single-crystalline silicon. This *unique device* will be utilized for ultra-precision dilatometric measurements of newly synthesized materials. In collaboration with Los Alamos National Laboratory (MST-NHMFL), the device is also planned to be used in the quasi-static 60T magnet for high-resolution thermodynamic determination of magnetic-field induced phase transitions.

Impact: Our recent efforts in the field of magnetic molecules has—within a short amount of time (1 year)—grown to an internationally competitive effort. This is reflected in the, so far, largest paramagnetic magnetic molecule $\{Mo_{72}Fe_{30}\}$ investigated, and the fact that we combine sample growth, cutting-edge experimental studies and theory within Ames Laboratory.

Interactions:
<u>U.S. National Laboratories:</u> M. Luban, P. Kögerler, F. Borsa, V. Dobrovitski, D.C. Johnston, J. Schmalian, Ames Laboratory; A. Migliori, N. Harrison, A. Lacerda, Los Alamos National Laboratory
<u>U.S. Universities</u>: Z. Fisk, J. Brooks, Florida State University, National High Magnetic Field Laboratory
<u>Non-U.S. Universities</u>: J. Schnack, D. Mentrup, Universität Osnabrück, Germany; A. Müller, Universität Bielefeld, Germany

Personnel:
R. Modler (group leader)

Recognition:

- Directors-funded Postdoctoral Fellow (Los Alamos National Laboratory, 1996-98)
- 3 invited talks since 08/98 at national and international conferences

Olson

Angle-resolved Photoemission

Description: This program uses synchrotron radiation based angle-resolved photoemission to study the electronic states of solids. The primary emphasis is on the states near the Fermi surface in systems including high temperature superconductors, heavy fermions, low dimensional materials and charge density wave compounds. The program also addresses broader questions of electronic structure in systems of interest within the Ames Lab, especially magneto-optic materials. The synchrotron radiation program involves a large number of collaborators from other institutions.

Highlights:

- Detailed, energy dependent, studies of the line shapes of the spectra of the oxy-chloride family of prototype high temperature superconductors. Strong photon matrix element effects are seen that must be considered in determining the true nature of the initial states.
- Detailed studies of conventional materials showing; a) true Fermi liquid behavior ($TiTe_2$), b) gapped states at the Fermi surface (dichalcogenide CDW systems), and c) reduced dimensionality (quasi-2D, e.g., $SmTe_3$, and quasi-1D, e.g., Mo bronzes). Fermi surface studies are showing when nesting is the source of the CDW.
- Studies of the degree of localization or hybridization of 4f and 5f levels in rare earth and uranium compounds, with an emphasis on heavy fermion materials.
- Photoemission studies of magneto-optical materials have complemented the lab based optical measurements.
- Studies of the MTe_5 and Bi_2Te_3 family of thermoelectrics have shown the temperature dependence of the electronic states responsible for the large thermoelectric efficiencies in these systems.

Impact: Our systematic studies across related systems have improved the understanding of the photoemission process in complex systems involving correlation, reduced dimensionality and gapped states at the Fermi surface. See also *Photoemission Studies of High-temperature Superconductors* by D. W. Lynch and C. G. Olson (Cambridge University Press 1999).

Interactions:
U.S. Universities: U. of Michigan (J. W. Allen), Michigan State U. (R. Liu), U. of Idaho (D. McIlroy)
Non-U.S. Universities: The Catholic Univ. of Korea (J.-S. Kang)
U.S. National Laboratories: LANL (A. J. Arko, J. Joyce), Ames Laboratory (D. W. Lynch, P. C. Canfield, B. N. Harmon, V. Antropov)

Personnel:
C.G. Olson (group leader), D. Brammeier (student)

Recognition:
- Fellow of the APS
- Synchrotron Radiation Center—Chair, Users' Advisory Committee
- Serve on several committees for ALS, APS, NSLS

Schmalian

Theory of Strongly Correlated Electron Systems

Description: The principal objective of our work is to develop theories for strongly interacting many body systems, with particular emphasis on interacting electrons in conducting, insulating, or superconducting states or in systems close to quantum phase transitions between these states. Non-equilibrium processes in correlated materials are also investigated. Strongly interacting electrons play an essential role in materials like high temperature superconductors, transition metal compounds, magnetic systems in general, organic conductors, heavy fermion systems and many other related materials. We use modern quantum field theoretical as well as numerical methods to investigate these systems. A close interaction with experimental groups in Ames and world wide is essential for a successful theoretical investigations of these systems.

Highlights:

- *pairing state in organic superconductors:* we predicted a pairing state for quasi 2-d organic superconductors which was experimentally confirmed by two groups.
- *quantum critical pairing in cuprates:* we developed a theory for the pairing state of incoherent fermions close to a quantum critical point, and applied our theory to cuprate superconductors
- *glassy dynamics in systems with charge-stripes:* we showed that a system which forms charge stripes due to a frustrating Coulomb interaction will undergo a transition to a "stripe glass" state at low temperatures, independent of whether the system is disordered or not.
- *defects in quantum critical metals:* we developed a theory for defect nucleated order and droplet dynamics in metallic systems close to a quantum critical point
- *domain disorder in magnetic multilayers:* we developed a theory for diffuse magnetic scattering due to domain disorder in magnetic multilayers

Impact: Our theoretical results have motivated several experimental as well as theoretical investigations throughout the world (for example in the groups of J. Singleton, Oxford; P. C. Hammel, LANL; C. P. Slichter, Urbana). Our theoretical predictions for the superconducting state in organic materials was later on experimentally confirmed and changed our understanding of these systems considerably. Our impact is also reflected in the recognition we received for our work listed below.

Interactions:
U.S. Universities: P. Goldbart, C. P. Slichter, Univ. of Illinois at Urbana-Champaign; P. G. Wolynes, UC San Diego, A. J. Millis, G. Kotliar, Rutgers Univ.; A. Chubukov, Univ. of Wisconsin at Madison,
Non-U.S. Universities: B. J. Hinkey, Univ. of Leeds, UK; J. Singleton, J. Betouras, Oxford Univ., UK, W. Hanke, Univ. of Wuerzburg; A. Rosch, Univ. Karlsruhe and K. H. Bennemann Univ. Berlin, Germany
U.S. National Laboratories: D. Pines, D. K. Morr, P. C. Hammel, LANL
Non-U.S. Laboratories: S. Langridge, S. Lovesey, Rutherford-Appleton Laboratory, UK

Personnel: J. Schmalian (group leader), H. Westfahl, Jr. (research associate), Sangwook Wu (student)

Recognition:
- Research Innovation Award of the Research Corporation (2000)
- ATLAS Fellow of St. Catherine's College, University of Oxford, UK (1999)
- 13 invited talks at national and international conferences since 1999

Shinar

Organic Semiconductor Materials and Devices

Description: The primary objective of this program is the study of the fundamental optical and electronic properties in p-conjugated materials and devices, mostly organic light-emitting devices (OLED's).

Highlights:

- Identification of the undesirable process of nonradiative quenching of the singlet excitons (SE's), which are the source of the photoluminescence (PL) and electroluminescence (EL) in these materials and devices, by trapped and free polarons, and quantification of this quenching mechanism.
- Identification of a similar SE quenching mechanism induced by triplet excitons (TE's), which may be even more significant in OLED's.
- Identification of trapping of injected electrons by negative charges, to form dianions, at the organic/cathode interface, by EL-detected magnetic resonance (ELDMR) of small molecular OLED's. This process is believed to strongly affect the efficiency of electron injection from the cathode to the organic layer.
- Discovery of sharp spikes at the turn-off of the EL induced by a bias pulse in blue 4,4'-bis(2,2'-diphenylvinyl)-1,1'-biphenyl (DPVBi)-OLED's. The spikes, with full-width-at-half-maximum durations of ~30 ns, are believed to result from the recombination of charges which accumulate at the interface between the electron- and hole-transporting layers. Detailed understanding of the spikes could lead to development of inexpensive pulsed light-sources based on such OLEDs.
- Development of various OLEDs: (a) Intense efficient UV-violet OLED's based on 4,4'-Bis(9-carbazolyl) bi-phenyl (CBP). We conducted a combinatorial study of matrix arrays of such OLEDs, in which the thickness of two layers was systematically varied across the array, and achieved an optimal radiance of ~0.2 W/cm^2 (equivalent to ~4 lumens/W at 555 nm) at 10 mA/cm^2, among the highest reported for UV-violet OLEDs. (b) Intense efficient UV-violet polymer OLED's based on poly(*N*-vinyl carbazole) (PVK). Although PVK has been used extensively as a polymer host for other emitting guest molecules, we have developed strong UV-violet OLED's in which this polymer is the emitting chromophore. (c) Strong blue OLEDs based on emission from an exciplex formed from *N,N'*-diphenyl-*N,N'*-bis(3-methylphenyl)-(1,1'-biphenyl)-4,4'-diamine (TPD) and 2-(4-Biphenylyl)-5-(4-*tert*-butylphenyl)-1,3,4-oxadiazole (Butyl-PBD). We are currently trying to optimize the emission and lifetime of these blue exciplex OLEDs. (d) Bright white multilayer small molecular OLED's.
- Development of integrated OLED/fluorescent chemical sensors.

Impact:

- The results on quenching of SE's by polarons and TE's demonstrate that these processes may be suppressing the efficiency of OLED's significantly.
- The development of various high-performance OLED's and the identification of the major processes which affect their performance, e.g., the spin-dependent formation of dianions at the organic-cathode interface (see above) should strongly impact the development of this emerging technology.
- The integration of OLEDs and optical chemical sensors could have far-reaching implications, such as providing the crucial first step towards miniature "on-chip"-compatible, very low-cost combinatorial sensor array systems, which would be a breakthrough as a technology and as a concept, completely reshaping the diagnostics and monitoring industries. It could be exploited in various environments, including *in vivo,* and even implanted, and include sensors of gases (O_2, CO, etc.) volatile organic compounds (toluene, benzene, etc.), biochemical compounds (glucose, gluthatione, etc.), and biological organisms (antibodies, antigens).

Interactions:
Technische Universität, Graz, Austria; eMagin Corp., Hopewell Junction, NY; Bell Labs, Lucent Technologies; University of Puerto Rico, San Juan; City University of Hong Kong; University of Korea, Seoul; Hebrew University, Jerusalem; National Academy of Sciences of the Ukraine, Kiev; Russian Academy of Sciences, Moscow; Univ of California, Los Angeles.

Personnel: J. Shinar (group leader); Postdoc: V. Savvateev (now at 3M); C.-H. Kim, L. Zou, G. Li, M.-K. Lee, K.-O. Cheon, B. Choudhury, and A. Naik.(graduate students); E. J. W. List, P. A. Lane, A. Qudah, A. Kadashchuk, E. Frankevich, I. Balberg, D. Davidov (visiting scientists).

Recognition:

- 17 invited talks at conferences and academic institutions since 10/1/98;
- 3 review articles (one published, one in press, and one in preparation) since 10/1/98;
- editor of three volumes (one published on fullerenes, one in press on OLEDs, and one in preparation on light-emitting polymers) since 10/1/98.

Soukoulis

Properties of Disordered Systems and Photonics

Description: Develop a theoretical understanding of the properties of disordered systems. Develop methods and understanding for the propagation of waves in periodic and random media. (Photonic Band Gaps (PBG), Random Lasers and Left-Handed Materials).

Highlights:

- A model to simulate the phenomenon of *random lasing* was developed. Good agreement with experiment and new predictions were obtained. In addition, our studies suggest that lasing in PBG materials with very low threshold can be realized experimentally.
- Theoretical modeling and design of *PBG materials* has led to the design of a 90-degree bend with 100% transmission. This has very important implications for optical communications. We have also a strong collaboration with European Labs to fabricate bends, splitters, optical switches, and resonators.
- We developed a *tight-binding* formulation of the EM wave propagation in *PBG structures*. The idea of the linear combination of atomic orbitals, well known from the study of electrons, was extended for the first time to EM waves. The formulation is used to understand the properties of defects and random PBGs.
- *Scaling properties* for the electrical conductivity in highly *anisotropic systems*, such as weakly coupled planes and weakly coupled lines, were studied. The behavior of the isotropic system was recovered once the proper scaling was completed. The *probability distribution of the conductance* at the mobility edge for both isotropic and anisotropic systems has been obtained. This is fundamentally important for understanding the transport properties of random materials.
- We began work on a new area of research, the so-called *left-handed (LH) materials*, which have both the dielectric constant and the magnetic permeability negative. LH materials have unique and potentially useful properties, as many basic EM phenomena, such as Doppler shift, Snell's law, and Cerenkov radiation are "reversed." We use our transfer-matrix and FDTD techniques to study their very interesting, challenging, and luckily unexplored properties.

Impact: The layer-by-layer PBG structure developed at Ames Lab has been the focus of many experimental groups all over the world in trying to fabricate PBGs, cavities and bends at the 1.5-micron scale. The random laser work not only explained a lot of the experiments, but made specific predictions that groups in the US and Europe verify and create random lasers with specific properties. Finally, our work on wave propagation on random media is well received and is used by many groups to determine the appropriate conditions for observing light localization. The number of citations (more than 500 per year), the number of invitations for talks on international conferences (more than 10 per year) and the requests to organize conferences and workshops gives extra dimension of the impact of our work.

Interactions:
International: E. Economou (Crete, Greece), P. Wolfle (Karlsruhe, Germany), Ad Lagendijk (Amsterdam, Netherlands), C. Weisbush (Paris, France
National: Hui Cao (Northwestern) on wave propagation in random media, random lasers and photonic crystals.
Industrial: Honeywell, 3M, and Boeing
Locally: K.-M. Ho, R. Biswas, G. Tuttle (EE), M. Tringides and G. Small (Chemistry).

Personnel: C. M. Soukoulis (Fellow, APS, group leader); Students: X. Jiang, S. Foteinopoulou, M. Ruehlaender, M. Agio (Pavia), Visitors: M. Kafesaki (Greece), P. Markos (Slovakia), E. N. Economou (Greece)

Recognitions:

- 12 invited talks last year at international conferences about our recent work on random lasers and photonic crystals
- Organized two international conferences last summer. Editor of both proceedings:
  - (i) "Wave propagation and electronic structure in disordered systems," a 60th birthday symposium to honor Prof. E. N. Economou, June 16-18, 2000 at Heraklion, Crete, Greece.
  - (ii) NATO ASI on "Photonic Crystals and Light Localization," June 19-30, 2000 at Limin Hersonnisou, Crete, Greece.
- Panel reviewer for the European Community and NSF to evaluate Physics and Engineering proposals.
- Published articles in the *Encyclopedia of Electrical and Electronics Engineering*, in the *McGraw-Hill Yearbook of Science and Technology*, and *Physicist's Desk Reference*.
- Over 400 citations per year, as stated in the *Science Citation Index*, for a total of 4,512 as of 2000.
- Named a 2000 recipient of the *LAS Outstanding Achievement in Research*. Named the 2001 recipient of the *ISU Outstanding Achievement in Research*.
- Our work on Photonic Band Gap Materials made both the *Energy 100 and Science 100 awards* of DOE.

Stassis

Neutron Scattering

Description: In this program elastic, inelastic, and polarized neutron scattering techniques are used to study the physical properties of condensed matter. Neutron scattering is actually the most powerful technique for the study of structural and magnetic phase transformations, magnetic and crystalline structure, elementary excitations (such as magnons, phonons), and quite generally the investigation of the physical properties of condensed matter in extreme environments. Our program includes the study of highly correlated electron systems, lattice dynamics of the high-temperature phases of various materials, and crystalline and magnetic phase transformations. Considerable effort is directed towards an understanding of the magnetic properties and lattice dynamics of high-T_c superconductors and related materials.

Highlights:

- *$RENi_2B_2C$ family of magnetic superconductors.* Our program established the magnetic structures of these compounds and explained various "anomalies" of these compounds and, in particular, the reentrant behavior of Ho compound at 5K. There are phonon modes in these compounds which are anomalous both below and above T_c. The behavior of these modes below T_c is related to the superconducting properties of these compounds and provides a measurement of the superconducitng gap.
- *Magnetic martensitic alloys.* It was established that in the presence of a magnetic field there is a large change in the elastic constants of Ni_2MnGa. A small but systematic hardening of the soft phonons was observed in a field of 70kG which may be due to the Zeeman splitting of the nesting feature of the Fermi surface responsible for the soft phonons. The premartensitic phase is considerably affected by the magnetic field.
- *Magnetoelastic compounds (RE)*$_5$ $(Si_xGe_{1-x})_4$. It was established that the structure of $Tb_5Si_2Ge_2$ compound evolves continuously between 950 and 10 K. The compound is orthorhombic at room temperature. A magnetic-crystallographic transformation takes place at approximately 100K.
- *Molecular magnets*. The magnetic spectrum obtained by measurements on a deuterated sample of a $\{Na_6V_6\}$ molecular magnet, which consists of two independent trimers of paramagnetic V^{4+} (s = 1/2) that interact via antiferromagnetic Heisenberg exchange is in good agreement with theoretical calculations.
- *Upgrade of the HB-1A Ames Laboratory Triple-Axis Spectrometer.* This instrument, installed at the HFIR of ORNL, is operated as a user facility jointly by the Ames Laboratory and ORNL scattering groups. Presently, the spectrometer is being modified and upgraded to accommodate the larger neutron beam (15 cm x 15 cm) that will become available after the upgrade of the HFIR. By horizontal and vertical focusing of the beam an increase of the intensity at the sample by a factor of approximately 2.5 has been estimated making the instrument the most powerful fixed-incident energy spectrometer in the United States.

Impact: Our results (such as the study of high-temperature phases of materials) have motivated many experimental and theoretical investigations. The HB-1A Ames Laboratory triple-axis spectrometer at the HFIR is hosting a large number of users from throughout the world.

Interactions:
Ames Laboratory: Goldman, Canfield, Lograsso, Gschneidner, Jr., Luban, Harmon, Ho.
U.S. Universities: U. of Missouri, Columbia U, Purdue U., SUNY Stony Brook
Non-U.S. Universities: U. of Barcelona (Spain); U. of Tokyo, Tohoku U., Kyoto U. (Japan); U. Bielefeld, U. Osnabück (Germany)
U.S. Laboratories: ANL, BNL, ORNL, NIST, LANL
Non-U.S. Laboratories: ILL, JAERI, Risø, Saclay, RIKEN, INSA, Commission of the European Communities Joint Research Center, Karlsruhe (Germany)
Industrial: NEC

Personnel: C. Stassis, D. Vaknin (group leaders); J. Zarestky (scientist permanently residing at Oak Ridge).

Recognition:

- Fellow of APS
- Co-recipient 1995 DOE Materials Science Award
- Several invited talks and lectures at national and international conferences
- 2 chapters in books in last two years
- Member of the executive committee and re-elected membership secretary, Neutron Scattering Society of America (NSSA)
- Member of the executive committee of SNS and HFIR users (SHUG)

Tringides

Control of Atomic Scale Growth on Surfaces

Description: We are interested in the characterization and control of atomic scale structures grown on clean surfaces. The control of the geometry and size of such structures requires an understanding of the key microscopic processes operating, such as surface diffusion, nucleation, island formation, etc. Such understanding is essential for the growth of custom-made materials of reduced dimensions with important applications in nanotechnology. For the experimental characterization, we employ two complementary techniques, Scanning Tunneling Microscopy (STM) and High Resolution LEED. We have strong interactions with the theory group to elucidate the physics behind the experimental results.

Highlights:

- *Discovery of self-organized, uniform height islands of Pb/Si(111) at low temperatures*. The grown islands have flat tops, steep edges and multiple step heights (5-,7-,9- etc., single steps). The origin of this highly unusual growth mode is most likely Quantum Size Effects (QSE), i.e., the quantization of the energy of the confined electrons in the islands.

- *Identification of the role of the metal/semiconductor interface in QSE*. By comparing the Pb uniform height islands grown on two different interfaces (Si(111)-(7x7) vs Si(111)-Pb-(√3x√3)), which have different electronic structures, we have shown the importance of charge transfer at the metal/semiconductor interface in selecting the preferred island height.

- *Correlation between real space and electronic structure of stepped Si(111)-(7x7).* With high resolution STM images and spectroscopic I-V measurements, we have shown correlations between changes in the atom position and local density-of-states for the step atoms on stepped Si(111), as a result of the step relaxation. These changes are in excellent agreement with the results of theoretical calculations by the theory group.

- *Development of High Resolution LEED as a real time, ultrafast acquisition method to study equilibrium dynamics on surfaces*. We have developed this new technique to study step fluctuations on Si(100) and W(430) surfaces at high temperatures and identified the microscopic mechanism (i.e., step diffusion, atom detachment, terrace diffusion, etc.) controlling the step fluctuations.

- *Determination of interlayer parameters for Ag/Ag(111)* . We separated the two contributions (i.e., barrier and prefactor) for the probability for an atom to move from a higher to a lower level for Ag/Ag(111). This probability is the key factor controlling whether growth is layer-by-layer or 3-dimensional in a system.

Impact: Our work on growth and surface diffusion is well recognized internationally, as evidenced by 19 invited talks over the last three years. In addition, we have organized two major international conferences on this topic Rhodes, Greece in 1996 and Prague in 2000. Recently our work on self-organized growth of Pb/Si(111) has initiated worldwide interest in this system by seven other research groups.

Interactions:
<u>Locally</u>: C. Z.Wang , K.-M. Ho, C. Olson, D. W. Lynch
<u>Outside</u>: Ed Conrad (Georgia Tech), Z. Chvoj (Academy of Sciences, Czech Republic) M. Henzler (Hannover, Germany), M. Hon-von-Hogen (Essen Germany), M. Jalochowski (Lublin, Poland) P. Argyrakis (Greece) M. Dudek (Wroclaw, Poland), K. Roos (Bradley).

Personnel: M. C. Tringides (group leader), M. Hupalo (Visiting Scientist), V. Yeh (PhD graduate student).

Recognition:

- 5 *Physical Review Letters* and one *Rapid Communication* in the last 3 years
- 19 invited talks at international meetings and institutions
- co-edited two conference books.

Vaknin

Neutron Scattering

Description: Our main objective is to employ and develop characterization techniques for novel materials using national facilities, such as research reactors, neutron spallation sources, and X-ray synchrotron sources. Our efforts have been devoted to two classes of systems—magnetic materials and thin organic films.

- Employing neutron scattering techniques we have been able to determine magnetic structures of new materials, characterize magnetic phase transitions, and magnetic excitations.
- We have been involved in the development of X-ray and neutron methods (instruments as well as methodology) to determine the structures of thin films (i.e., spread monolayers at the air-water interface, thin polymer films etc.). In particular, we have recently constructed a novel X-ray liquid surface diffractometer at the APS at Argonne National Laboratory to complement neutron diffraction studies.

Highlights

- *Magnetic clusters in $BaCuO_{2+x}$.* Employing neutron scattering diffraction techniques we were able to identify a large single crystal in a melt, and study its unique magnetic properties. $BaCuO_{2+x}$ has 90 Cu^{2+} spins per unit cell that form two kinds of clusters; ring-like Cu6 and sphere-like Cu10. Neutron diffraction studies under applied magnetic fields allowed detailed characterization of this unique system.
- *Magnetic properties as probes for studying fast ionic conductors.* The family of Li phosphates $Li_xM_y(PO)_z$ (M = Fe, Mn, Ni), are being examined as possible fast ionic conductors for use as rechargeable batteries. Neutron diffraction studies have been conducted to determine the chemical and magnetic structures of these systems.
- *Magnetoelectric effect materials.* Puzzling phenomena observed in the magnetoelectric effect exhibited by single crystals $LiNiPO_4$ and $LiCoPO_4$ can mow be explained by our neutron diffraction results that reveal a unique commensurate-incommensurate magnetic transition, and canted moments in the ordered antiferromagnetic state of these two systems, respectively.
- *Cuprates and high Tc superconductors*. Neutron diffraction studies of a superconducting single crystal $YBa_2Cu_3O_{7-x}$ under applied magnetic field show weak peaks that might indicate that vortex cores in the superconducting state are antiferromagnetic. Other studies of the insulating parent materials $Ca_2Cu_2O_2Cl_2$, $Sr_2Cu_2O_2Br_2$ reveal the well known AF in-plane ordering of Cu^{2+} spins (S = 1/2) but with spiral stacking along the planes' normal.
- *Liquid surfaces diffractometer at the APS*. We have successfully installed and commissioned an X-ray Liquid Surfaces Diffractometer at the Advanced Photon Source at Argonne National Laboratory on the 6-ID beamline of the Midwestern Universities CAT. This is a unique instrument that allows studies of liquid surfaces using X-ray energies in the range of 4 to 40 keV. In the commissioning process, the diffractometer has been employed to investigate a variety of systems.
- *Molecular magnets at the air/water interface*. We have been able to form a single layer of the AF Cr8 molecule Cr_8O_4 $(O_2CPh)_{16}$ at the air/water interface and characterize its structure using the Liquid-Surfaces Diffractometer at the synchrotron.
- *Anomalous x-ray reflectivity.* Performing reflectivities at and off the absorption edge of Ba^{2+} (anomalous X-ray Reflectivity) we have been able to characterize the partial layer of bound Ba^{2+} ions to a Langmuir monolayer. These studies are of importance for the understanding of biological phenomena in the vicinity of cell membranes, and help in the development of artificial bio-membranes.

Impact: Our research is multidisciplinary in nature, and draws the attention of physicists, chemists, and biologists, which are interested in using the tools that we developed to further understand their systems. Our magnetic studies as well as those of thin membranes have also stimulated numerous theoretical papers.

Interactions

Local: Physics Department, Chemistry Department, and Chemical Engineering Department.
US Universities: Univ. Pennsylvania; Kent State, Ohio.
Non-US Universities: Univ. Leipzig Germany; Univ. Geneva, Switzerland; Univ. Bilbau, Spain; Campinas, Brazil; Univ. Quebec, Canada.
US National Laboratories: Oak Ridge National Laboratory, NIST, Argonne National Laboratory, Brookhaven National Laboratory.

Personnel

David Vaknin (group leader), Michael Kelley and J. McManus (students)

Recognition

- Member of the executive committee of SNS and HFIR Users (SHUG).

Wang

Surfaces–Theory

Description: The principal objective of the program is the study of fundamental interactions in solids and on solid surfaces through atomistic modeling and computer simulations. Our research in the past three years includes: (i) atomic process and activation energetics of small structures (adatoms, addimers, and small clusters) on semiconductor surfaces (partially in collaboration with M. G. Lagally's experimental group at University of Wisconsin, Madison); (ii) atomic and electronic structure of stepped silicon surfaces and electronic structure of metal-semiconductor interfaces (in collaboration with M. Tringides experimental group at Ames Laboratory); (iii) structure and property of semiconductor clusters (in collaboration with M. F. Jarrold experimental group at Northwestern University); (iv) laser ablation of semiconductor surfaces (in collaboration with P. Molian's experimental group at Iowa State University). We have developed several environment-dependent tight-binding potentials for accurate and faster molecular dynamics simulations and a tight-binding/genetic-algorithm scheme for efficient atomistic structure optimizations. We also had our state-of-the-art first-principles density functional code implemented on the massively parallel computers (IBM-SP3 and Cray-T3E) at DOE's NERSC for large scale ab initio calculations.

Highlights:

- *Silicon ad-dimer diffusion on Si(001*). Using a combination approach of tight-binding molecular dynamics simulation and ab initio calculations, we discovered several low-energy diffusion pathways that were missed in previous studies.
- *Diffusion and Intermixing of SiGe addimer on Si(001*). Our calculations help experiment to identify the mixed SiGe addimer on the Si(001). Our calculations also provided detailed information about the atomic process of diffusion and intermixing of mixed SiGe addimer on Si(001).
- *Atomic and Electronic Structure of Stepped Si(111)-(7x7) Surface*. Our studies reveal several new surface bands induced by the step on the Si(111)-(7x7) surface and identify the origin of these new bands.
- *Structure and Properties of Si Clusters:* We have determined the lowest-energy structures of the medium-sized silicon clusters (n=11-20) and studied their properties.
- *Nanoscale Pb Island on Si(111*). Our study shows that the stable height of the Pb island on Si(111) surface is dependent on the structure and hence charge transfer at the metal/semiconductor interface.
- *Si Cluster on Si(111)-(7x7) Surface*. A systematic search for the lowest-energy structure of silicon clusters (n=4-14) on Si(111)-(7x7) surface has been performed using the TB/GA .
- *Atomistic Simulation of laser-Induced Graphitization on a Diamond (111) Surface*. Our simulation results suggest a non-thermal mechanism using femtosecond laser pulse, which leads to a layer-by-layer graphitization of diamond (111) surface and results in a clean diamond surface after laser ablation.
- *Reconstruction of the Mo(100) Surface by Tight-Binding Calculations*. We have developed an accurate environment-dependent tight-binding potential for Mo which describes well the properties of Mo including the complex reconstruction of the Mo(001) surface.

Impact: Our results on the adatom and addimer diffusion on Si(001) surface provide the most accurate and detailed picture about the atomic process at the the initial stage of epitaxial growth of Si and Ge on Si(001). The tight-binding molecular dynamics pioneered by our group has been adopted by many research groups in the U.S. and other countries for material simulations. (E. Kaxiras' group, Harvard; D. Pettifor's group, Oxford, as examples)

Interactions:
M. G. Lagally group (experiment) of Univ of Wisconsin, Madison; M. Tringides experimental group at Ames Laboratory; M. F. Jarrold experimental group at Northwestern University; P. Molian experimental group at Iowa State University.

Personnel: C. Z. Wang and K. M. Ho (principal investigators), Z. Y. Lu (post doc.), B. Liu (student)

Recognition:

- 1 invited talk at international conferences and a physics colloquium in the last two years.

Zarestky

Neutron Scattering

Description: The objective of our program is to employ neutron scattering techniques in the study of novel materials. Neutron scattering has been used to investigate structural and magnetic phases and their respective transitions as well as the lattice dynamics of these materials. The principal instrument used by this program is the Ames Laboratory spectrometer (HB1A) at the High Flux Research Reactor (HFIR) at Oak Ridge National Laboratory (ORNL). This machine was designed, built, and is presently operated as a users facility by Ames Laboratory and the ORNL neutron scattering groups. Presently the HFIR beam lines are undergoing a major upgrade that includes installing larger beam tubes. In order to take advantage of the increased beam size the HB1A spectrometer is being upgraded from the first monochromator position to the sample position and eventually to the detector/analyzer system. With this upgrade, the flux at the sample position will be comparable to that available at the best ILL instruments.

Highlights:

- *Phonon mode coupling in superconducting RE*Ni_2B_2C. Our studies show that the strong phonon softening above T_c is caused by mode coupling, and the dramatic phonon profile changes below T_c are consistent with the response expected in a BCS superconductor.
- *Soft phonons in magneto-mechanical Ni-Mn-Ga alloys.* Measurements of the TA_2 branch show that the wavevector of the soft phonon and its temperature dependence, vary with sample composition.
- *Magnetic energy levels of the molecular magnet* $\{Na_6V_6\}$. Preliminary neutron scattering experiments are in reasonable agreement with theoretically predicted energy levels.
- *Magnetic properties of fast ionic conductors.* Studies using neutron diffraction techniques have been conducted to determine the chemical and magnetic structures of a family of Li phosphates $Li_xM_y(PO)_z$ (M = Fe, Mn, Ni). These systems have potential as possible fast ionic conductors for use in rechargeable batteries.
- *High Tc superconductors*. Weak peaks observed by neutron diffraction on a superconducting single crystal of $YBa_2Cu_3O_{7-x}$ under applied magnetic field may indicate that vortex cores in the superconducting state are antiferromagnetic.
- *Magnetoelectric Effect Materials.* Neutron diffraction measurements on single crystals of $LiNiPO_4$ and $LiCoPO_4$ have explained observed magnetoelectric effect behavior.

Impact: Because of the fundamental nature of our research the results are of interest to many investigators, from those producing the samples all the way to those using the materials in applications. Our results are also used by theorists studying these and related materials and phenomena. The HB1A spectrometer has proven itself to be world class instrument in providing contamination free flux on the sample and low background. It has been used by a large number of users from the U.S., Europe, Canada, and Japan.

Interactions:

Local (Ames Laboratory / ISU): Physics Department; Metallurgy Department.

US Universities: Univ. of Tennessee, Knoxville.

Non-US Universities: Univ. of Tokyo, Japan; Univ. Bilbau, Spain; Campinas, Brazil; Universitat de Barcelona, Barcelona, Spain; Universidad Nacional del Centro de la Provincia, Buenos Aires, Argentina; Univ. Villeurbanne, France.

US National Laboratories: Oak Ridge National Laboratory, Argonne National Laboratory, Brookhaven National Laboratory, Los Alamos National Laboratory.

Non-US Laboratories: Institut für Festkörperforschung, Jülich, Germany; Institute of Physical and Chemical Research (RIKEN), Japan; Institut Laue-Langevin, Grenoble, France; Oxford University, U.K., Commission of the European Communities Joint Research Center.

Personnel:

J. L. Zarestky (scientist, Ames Laboratory / ISU Neutron Scattering group at ORNL)

Recognition:

- Co-recipient 1995 DOE Materials Science Award
- CG1 HFIR Guide Hall Cold-Triple-Axis Spectrometer design team member